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JOINT APPENDIX

# United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA CIRCUIT

APPEAL NO. 13,566

ROBERT C. WATSON, COMMISSIONER OF PATENTS,
APPELLANT

*v.*

FREDERICK C. BERSWORTH AND THE DOW CHEMICAL COMPANY, APPELLEES

*APPEAL FROM THE UNITED STATES DISTRICT COURT FOR THE DISTRICT OF COLUMBIA*

United States Court of Appeals
For the
District of Columbia Circuit
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Joseph W. Stewart
CLERK

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# United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA CIRCUIT

APPEAL NO. 13,566

ROBERT C. WATSON, COMMISSIONER OF PATENTS,
APPELLANT

*v.*

FREDERICK C. BERSWORTH AND THE DOW
CHEMICAL COMPANY, APPELLEES

*APPEAL FROM THE UNITED STATES DISTRICT COURT FOR THE DISTRICT OF COLUMBIA*

JOINT APPENDIX

 United States District Court for the District of Columbia

Civil Action No. 1637—54

FREDERICK C. BERSWORTH, FRAMINGHAM,
MASSACHUSETTS, PLAINTIFF,

*v.*

ROBERT C. WATSON, COMMISSIONER OF PATENTS,
WASHINGTON, D. C., DEFENDANT

TO OBTAIN PATENT UNDER 35 U. S. C. § 145

*Complaint*

Filed April 19, 1954. Harry M. Hull, Clerk.

Frederick C. Bersworth, the Plaintiff herein, brings this Complaint against Robert C. Watson, Commissioner of Patents, Defendant, and alleges that:

1. The Plaintiff herein, Frederick C. Bersworth, is a resident of Framingham, in Middlesex County, in the State of Massachusetts.

2. The Defendant, Robert C. Watson, is the United States Commissioner of Patents, and is officially a resident of Washington, in the District of Columbia, within the jurisdiction of this Court.

3. This action is brought under the provisions of U. S. C., Title 35, Section 145; 66 Stat. 803.

4. On or about the 12th day of March 1951 Frederick C. Bersworth, the Plaintiff herein, executed an application for Letters Patent for a composition of matter, "Hydroxy Derivatives of Alkylene Polyamines", invented by him.

5. On or about the 13th day of March 1951, the said Plaintiff duly made application in writing in
117 the United States Patent Office for the grant of Letters Patent to him for the composition of matter described and claimed in the said patent application, which application was given Serial No. 215,380.

6. Said application for Letters Patent contains the claims to which Plaintiff believes himself entitled, said claims being numbered 1 through 4.

7. Thereafter, said application for Letters Patent was examined by the Examiner in charge thereof, who, on the 22nd day of January 1952 finally rejected the said claims; on the 21st day of July 1952 an appeal was duly taken to the Board of Appeals of the United States Patent Office, and, on the 18th day of February 1954 said Board of Appeals rendered its decision, affirming the rejection of the claims by the Examiner.

8. Plaintiff alleges that the decision of the Board of Appeals adjudging the Plaintiff not entitled to Letters Patent for the composition recited in said claims 1 through 4 was erroneous and contrary to law.

9. Plaintiff further alleges that no appeal has been taken by him to the United States Court of Customs and Patent Appeals from said refusal of the Commissioner of Patents to issue said Letters Patent, and that the Complaint herein is filed within the sixty (60) day limitation, as extended by Rule 304 of the United States Patent Office Rules of Practice, imposed upon filing appeal from the decision of the Board of Appeals refusing to allow the aforesaid claims and issue Letters Patent on said composition.

118 10. Plaintiff makes profert of a certified copy of the aforesaid application for Letters Patent, Serial No. 215,380, and all proceedings and papers in the file thereof, together with certified copies of the patents forming the basis for the aforesaid decision refusing to allow the claims and issue Letters Patent on said invention, said certified copies to be produced as and when this Honorable Court shall direct.

Wherefore, Plaintiff respectfully prays as follows:

First: For a decree that Plaintiff is entitled to receive Letters Patent for the composition shown and described in the aforesaid application, Serial No. 215,380, in due form of law as prescribed by the Statutes.

Second: For a decree pursuant to U. S. C., Title 35, Section 145, authorizing the Commissioner of Patents to issue Letters Patent on said composition, in due form of law and as prescribed by the Statutes.

Third: That Plaintiff have such other and further relief as the nature of the case may admit or require and as may be just and equitable.

Respectfully,

Ellsworth H. Mosher,

ELLSWORTH H. MOSHER,
*Attorney for Plaintiff,*
*Munsey Building, Washington 4, D. C.*
THOMAS B. GRAHAM,
*405 Lexington Avenue, New York 17, N. Y.,*
DARBY & DARBY,
*405 Lexington Avenue, New York 17, N. Y.,*
*Of Counsel.*

119 United States District Court for the District of Columbia

Civil Action No. 1637-54

FREDERICK C. BERSWORTH, PLAINTIFF

*v.*

ROBERT C. WATSON, COMMISSIONER OF PATENTS, DEFENDANT

E. L. REYNOLDS,
*Solicitor, U. S. Patent Office,*
*Washington 25, D. C.,*
*Attorney for Defendant.*

*Answer to the Complaint*

Filed April 29, 1954. Harry M. Hull, Clerk.

*To the Honorable Judges of the United States District Court for the District of Columbia.*

1, 2, 3, 4, 5, 6, 7. The defendant admits the allegations of paragraphs 1 to 7, inclusive, of the complaint.

8. He denies the allegations of paragraph 8.

9, 10. He admits the allegations of paragraphs 9 and 10.

Further answering, the defendant states that plaintiff is not entitled to a patent containing any of the claims referred to in the complaint for the reasons given and in view of the references cited in the Examiner's statement and answer to the plaintiff's appeal, and in the decision of the Board of Appeals. Profert of copies of the said state-
120 ment, decision, and references is hereby made.

Respectfully submitted,

E. L. REYNOLDS,
*Solicitor, U. S. Patent Office,*
*Attorney for Defendant.*

APRIL 27, 1954.

I hereby certify that a copy of the foregoing Answer to the Complaint was mailed today to the attorney for the plaintiff, Mr. Ellsworth H. Mosher, Munsey Building, Washington, D. C.

E. L. REYNOLDS,
*Solicitor.*

121 In the United States District Court for the District of Columbia

Civil Action No. 1637-'54

FREDERICK C. BERSWORTH, PLAINTIFF,

*v.*

ROBERT C. WATSON, COMMISSIONER OF PATENTS, DEFENDANT

*Stipulation to Join Additional Party as Co-Plaintiff*

Filed February 17, 1955. Harry M. Hull, Clerk

It is stipulated by and between the undersigned attorneys for plaintiff and defendant, respectively,

that The Dow Chemical Company, a corporation of Delaware, having its offices and principal place of business in Midland, Michigan, has acquired the entire right, title and interest in and to the U. S. patent application Serial No. 215,380 involved in this action, and in and to the invention represented thereby, as witnessed by a Deed of Assignment recorded in the United States Patent Office at Liber I-243, page 496, on the 13th day of December, 1954, and said attorneys therefore respectfuhy move the Court to make The Dow Chemical Company an additional party co-plaintiff to this action.

E. L. Reynolds,
E. L. REYNOLDS,
*Solicitor, U. S. Patent Office,*
*Attorney for Defendant.*

Ellsworth H. Mosher,
ELLSWORTH H. MOSHER,
*Attorney for Plaintiff.*

FEBRUARY 16, 1955.
THOMAS B. GRAHAM,
DARBY & DARBY
*405 Lexington Avenue,*
*New York, 17, N. Y.,*
*Of Counsel.*

122 In the United States District Court for the District of Columbia

Civil Action No. 1637—'54

FREDERICK C. BERSWORTH, PLAINTIFF

*v.*

ROBERT C. WATSON, COMMISSIONER OF PATENTS,

DEFENDENT

*Order of Joinder of Additional Party as Co-Plaintiff*

Filed February 21, 1955. Harry M. Hull, Clerk.

Upon consideration of a stipulation between the attorneys for plaintiff and defendant, respectively, and upon their joint motion, it is this day hereby

Ordered that The Dow Chemical Company, a corporation of Delaware, with offices and principal place of business in Midland, Michigan, be brought upon the record as a party co-plaintiff to this action in addition to the original party plaintiff Frederick C. Bersworth.

R. B. KEECH,
*U. S. District Judge.*

FEBRUARY 21, 1955.

123 In the United States District Court for the District of Columbia

Civil Action No. 1637–'54

FREDERICK C. BERSWORTH AND THE DOW CHEMICAL COMPANY, PLAINTIFFS

*v.*

ROBERT C. WATSON, COMMISSIONER OF PATENTS, DEFENDANT

*Stipulation Re Voluntary Dismissal of a Portion of Civil Action Under F. R. C. P. 41 (a) (1) (ii)*

Now come the parties, by their attorneys, and stipulate that so much of this civil action as relates to nonelected species claims 1 and 3 of plaintiffs' patent application Serial No. 215,380 here in suit, which claims correspond respectively to claims 1 and 2 of patent application Serial No. 496,338 filed March 23, 1955 and currently pending in the U. S. Patent Office, may be and hereby is dismissed without prejudice under F. R. C. P. 41 (a) (1) (ii).

C. W. MOORE,
*Solicitor of U. S. Patent Office,*
*Attorney for Defendant.*

MARCH 26, 1956.

Ellsworth H. Mosher,
ELLSWORTH H. MOSHER,
*Attorney for Plaintiffs.*

MARCH 12, 1956.

United States District Court for the
District of Columbia

Civil Action No. 1637—'54

FREDERICK C. BERSWORTH, FRAMINGHAM, MASSACHUSETTS, AND THE DOW CHEMICAL COMPANY, MIDLAND, MICHIGAN, PLAINTIFF

*v.*

ROBERT C. WATSON, COMMISSIONER OF PATENTS, WASHINGTON, D. C., DEFENDANT

WASHINGTON, D. C., *April 17, 1956.*

The above-entitled matter came on for hearing before the Honorable Robert N. Wilkin, in Courtroom 14, United States District Court for the District of Columbia, 3rd and Constitution Avenue, Northwest, Washington, D. C., on April 17th, 1956, commencing at 10:00 o'clock a. m.

Appearances: On behalf of Plaintiff: Thomas B. Graham, Esq., and Ellsworth H. Mosher, Esq. On behalf of Defendant: Joseph Schimmel, Esq.

PROCEEDINGS

Mr. MOSHER. Good morning, your Honor. This is a Civil case under 35, section 145 of the U. S. Code, against the Commissioner of Patents, seeking an order authorizing the issuance of a patent to Plaintiffs.

My colleague, Mr. Thomas Graham, of New York City, with your permission, will conduct the case. It so happens he is a member of the bar of this Court.

The COURT. Very well.

Mr. GRAHAM. May it please the Court, this is a fairly common type of case in this jurisdiction, and its characteristic is that a plaintiff who is dissatisfied with the Patent Office's decision comes in seeking his patent, because he has been denied by the Patent Office.

The COURT. I am quite familiar with the procedure.

Mr. GRAHAM. This is a bit unusual in that the Patent Office has conceded we have invention, but they still refuse to grant a patent for some procedural reasons.

Now, the subject matter of the invention is an organic chemical compound. It is fairly complicated organic chemistry, but in as much as invention has been conceded it will be unnecessary for us to consider details of the chemistry; we can confine our attention purely to the procedural matters involved.

For the background of what we are to look at, it will be necessary to take a brief look at the chemistry first.

Now, there is only a single claim before the Court. It is the elected species, and we will come to considering that in detail in a few moments. The compound is in a class of materials which we call chelating agents. That is a very picturesque word derived from the Greek word "chelos", meaning claw.

It is a type of organic compound which has characteristic structural units on it arranged spatially so that they will react with meal ions in solution and hold those ions in a form such that chemically they are no longer present in the solution.

Now, most people can go through life without hearing of chelating agents. In fact, many chemists can. But it is a very common and useful industrial phenomenon, and perhaps it will be good at this point to look a little bit at the chemistry that we have.

The compounds that we have have a characteristic structure which is a nitrogen, then a central hydrocarbon structure carrying another nitrogen on the end. That is the starting nucleus we have to consider. We have acetic groups, and that is the char . eristic structural entity vhich runs throughout the entire class of compounds we are studying.

Chemically, it has a complicated name, but it is this structural entity.

Now, we have before the Court a compound in which additional groups have been placed in these positions. [Indicating on blackboard.] That is, the upper corners of the molecules, to change the symmetry of it and enhance its chelating and its chemical usefulness properties in that fashion.

That is what we will be looking at essentially, throughout our consideration of the case. That was a concept which was developed by Doctor Bersworth. He spent his entire adult life studying that chemistry, and, in effect, he has become Mr. Chelation to the chemical industry. He built up a very prosperous little business on the manufacture of compounds of that type. And in December of 1954, he sold the entire business to the Dow Chemical Company, and Doctor Bersworth is now a consultant for Dow, and at the present moment is completing a grand tour of the Dow Chemical establishments in the United States.

He has spoken his piece in the patent application and will not be here this morning.

Now, commercially, these compounds ar useful in many places. In fact, wherever water is used it must be treated as chemical material because there will always be contamination in water with traces of metal ions, calcium, magnesium and very often, traces of iron.

When you have that condition, these traces of metals will affect the process which is being carried on in a water medium, and by the addition of this chelating agent you can render the trace metal ions effectively not present in the water. They will not enter in the process you are doing.

In fact, our shirt manufacturers would be unable to turn out nice white shirts without chelating agents

because by adding it to the water it is possible to tie up the calcium, magnesium and iron present and wash the shirts in nice pure soap and get a clean result.

Now, the specific claim we have before the Court, again, that will be looked at in detail. The formula for it is written in this form, with the notation used in the application. [Indicating on blackboard.] It has been made more specific. And we have used different letters in this position [indicating] and up here we have used letters A, but that is merely in the consideration of the general type compound we have used that formula.

The specific exact compound, or the prefered specific compound which is covered by the claim is one which has an acid group in each of these positions [indicating]. Acid group being an acetic group. And it has what we have called in the application an ethanol group in this location [indicating on blackboard].

Now, this is all conventional chemical notation, and we have laid it out in exhibit form so that we can refer to these things by exhibit number as we proceed.

Now, I say the Patent Office has admitted that we have an unusual compound which ought to be patentable, in fact, is inventive, but they have some difficulty with procedural matters by maintaining that the subject matter was disclosed in the Patent Office at an early date and was not claimed. And accordingly, a couple of our own earlier patents are anticipation of it.

We have demonstrated within the Patent Office where this subject matter has been carried continuously in a series of divisional and continuing applications made necessary by requirements of the Patent

Office. So that we are here today with an application which is a continuation of an earlier one asserting these claims.

But the Patent Office says that one of the intermediate applications on which we depend for this continuity, if it is sufficient to support our claim, gives them the right then to read a certain other reference which is in the record to find an anticipation of this claim.

So, basically, our issue before the Court this morning is the patentability of that compound on a double-barreled argument from the Patent Office. Either we have failed to maintain continuity or the compound is anticipated.

That summarizes the outline of what we will present. I will hold the exhibits and offer them as the presentation of the case goes along. I have prepared a trial memorandum in which the case has been made out in some detail with a diagram to simplify the problem of following continuity here.

The diagram is in large form because it gets a little difficult to maintain four or five or six digit serial numbers in the mind as you read this subject matter.

Mr. SCHIMMEL. If the Court please, I would like to take exception to the statement made by Mr. Graham that the refusal of the Patent Office in this case is based upon procedural aspects. Refusal of the Patent Office is based upon the specific provisions of the statute, and I would like to call your Honor's attention to those provisions, if I may. Section 102 is the basic statute setting forth the novelty aspects warranting the grant of a patent. And subsection (b) states that "The person shall be entitled to a patent unless the invention was patented or described in a printed publication in this or any foreign country

more than one year prior to the date of the application for patent in the United States."

And subsection C states, "He will be entitled to the patent unless he has abandoned the invention."

Now, our position and the position of the Patent Office throughout the prosecution of the present application is that two of the Plaintiff's, Bersworth, patents, published the subject matter which is sought to be protected in this case. And those patents were granted and published more than one year before any effective date to which he is entitled.

Therefore, he would be barred under the publication provision of the statute.

Secondly, we say the same patents disclose a compound within the scope of the broad subject matter that is being relied on as a basis for the right to claim this specific compound in the present case.

That subject matter was abandoned to the public by Doctor Bersworth's failure to continue the disclosure of that subject matter in pending applications before the Patent Office. That is, the doctrine of continuity which will be urged here this morning requires the party to continually keep before the Patent Office in a pending application, the disclosure upon which he is to rely subsequently in order to entitle him to the benefit of the filing date of that early application.

Our position is that the references and evidence will show that that has not been done in this case. More than one year elapsed between the disclosure in Bersworth's earlier patent, and the first application which we say represented that identical subject matter to the Patent Office.

Under the law, that is a conclusive abandonment of that particular subject matter, and therefore, Plaintiff is not entitled to recapture it at this late date.

Those are the issues which we feel are before the Court this morning. And I would like to hand up to your Honor the folder which we will use as our official exhibit, and refer to the decision of the Board of Appeals, which is item E in that folder; on page two of that decision in the last paragraph on the page, there appears this statement: "There appears to be no dispute on the fact that these patents disclose compounds which anticipate the appealed claims."

The argument for patentability is that these patents are not proper reference by virtue of the fact that appellant had pending before the Patent Office, this subject matter, prior to the effective dates of these patents. Appellant traces a chain of continuity back to the parent patent 2,407,645, filed June 21, 1943. He states that the filing of applications, serial numbers 39377 and 39378, which covered the genus and species of the instant application prior to issue of patent 2,524,218, completely negatives abandonment of the invention now claimed in the present application, which is a continuation of serial number 39377.

There we have the aspect of no dispute as to whether or not there is anticipation of the subject matter. We have the aspect that abandonment is the reason for which the Patent Office has refused the claims in this particular case, and we have the reliance by the Plaintiff here on a genus, a generic disclosure in the parent application on which he alleges he is entitled support in this specific claim he has indicated on the blackboard. We assert he is not entitled to do that under the prevailing authorities, and that the refusal of the Patent Office is proper under the law.

Mr. GRAHAM. May we proceed now?

The COURT. You may.

Mr. GRAHAM. As a matter of interest, not as an exhibit, I will offer the Court an opportunity to inspect the commercial form in which this compound is sold. You might mark that as an exhibit for identification. It is a 34 per cent solution, aqueous solution of the compound we are considering, and it is the common form, which is sold industrially, principally because the material is used in the water solution and they merely sell it in concentrated solution. In the dry form it looks like powdered sugar and is an innocuous appearing material.

At this stage I would like also to offer up and have marked as our first exhibit for identification, a collection of references which have been cited and discussed in the record of the Patent Office, and we will make much reference to this during the next few minutes of the proceeding.

The CLERK. They will be marked for identification as Plaintiff's one and two.

Mr. GRAHAM. At this point I would like to call as my first witness, Doctor Arthur Martell.

Thereupon Doctor ARTHUR E. MARTELL was called as a witness by counsel for Plaintiff, and having been first duly sworn, was examined, and testified as follows:

Mr. GRAHAM. Doctor Martell is a man who worked a long time with Bersworth. He is quite familiar with this case and is an expert in the field of Chelation. And that is the purpose of calling him in.

The COURT. How do you spell that word you used last?

Mr. GRAHAM. Chelate, C-H-E-L-A-T-E.

Direct examination by MR. GRAHAM:

Q. Will you state your name, please?

A. Arthur E. Martell.

Q. What is your address?

A. My address is Maynard street, Northboro, Massachusetts.

Q. And your occupation?

A. Professor of Chemistry at Clark University, Worcester, Massachusetts.

Q. Will you describe briefly your education and experience and make reference briefly to the publications and the like which would qualify you as expert?

A. I have received my Phd. in chemistry at New York University in 1941. From that time on I have been a teacher of chemistry in colleges and universities. I was appointed to professor at Clark University in 1950. I have written a book on metal chelate compounds. I have also been research fellow at the University of California for one year, where I investigated these substances. I have approximately fifty publications in the literature on the properties of compounds of this type.

Q. Do I understand you are a member of the International Union of Pure and Applied Chemistry?

A. I am a member of the subcommittee of this Union on the stability of metal chelate compounds.

Q. This is the committee that devotes its time to the specific study of compounds of this type?

A. Yes.

Q. Are you familiar with the subject matter and details of this case before the Court?

A. I am.

Q. Are you familiar with the rules of organic chemical nomenclature?

A. I am.

Q. Now, for most organic compounds, are these rules of organic nomenclature standardized?

A. They are.

Q. Now, in this standardized system is it generally accepted by chemists?

A. Yes.

Q. When a new compound has been made, can that new compound be named by using these rules of nomenclature?

A. Yes.

Q. Does organic chemical nomenclature then give a skilled chemist a word picture of the structural formula of an organic chemical compound?

A. It does.

Q. Now, taking the obverse of that question, does the structural formula, for example, that one on the blackboard, give a skilled chemist a word name for an organic compound?

A. Yes.

Q. Is it correct for us to say that organic chemical nomenclature constitutes an agreed upon word amongst chemists for describing chemical compounds?

A. Yes.

Q. Is it correct then to say that this word code is used to convey a description of organic chemical structure so that chemists can reproduce that structure?

A. It is.

Q. It is correct for us to say then that an organic formula written in structural form is the equivalent of an organic name derived from these rules of nomenclature?

A. Yes.

Mr. GRAHAM. At this point, I believe we can start tracing the continuity of the subject matter we have before the Court.

The COURT. When you use the word "name" in the questions you asked, just what do you mean, the popular name?

Mr. GRAHAM. No; that is something else.

The COURT. That is the trade name?

Mr. GRAHAM. That is something else again. Names of substances can be trivial or popular names. For example, the name for this term [indicating on blackboard] is water. A chemical term for it would be hydrogenoxide. A common name for table salt is "salt." Chemically, it is sodium chloride.

Now, when the chemistry of organic compounds began to be developed it was found very quickly these trivial names were exhausted, because there are so many compounds, and the permutations and combinations available in structures became so complicated that a system had to be developed for describing them, and that is this system of nomenclature we are talking about.

The COURT. I understand.

Mr. GRAHAM. On common alcohol, or ethyl alcohol as we call it, the systematic organic name for it would be ethanol. And as the compound became more complex it became impossible to describe them in words of three or four or even five syllables.

Many, many words have to be strung together with numerical designations to indicate where these groups go.

The COURT. The word you used in your question is the name for this combination, and is the name you have given in your complaint, is that right.

Mr. GRAHAM. Yes. This compound can be named in several ways; each of them would be a definitive organic name. The name that is on the label of the solution you have before you is a trade mark under which the material is sold. In the patent application we are talking about, this compound is called diethanol, meaning two ethanol groups, ethylene, referring to this central portion, diamine, referring to the nitrogens, diacetic acids, meaning the two acids.

That name can be dressed up to be more definitive by calling these things hydroxyethyl groups. And the name can be written in several alternative forms, each of which is definitive of this structure.

The COURT. I understand.

Mr. GRAHAM. Now, in tracing this continuity, it would be convenient for us to refer to the blueprint which has been incorporated in the trial memorandum. And a favorable starting point would be with the case which has been labeled case A. A 1 that is one which carries the full designation of the U. S. Patent case 2,524,218. It is one of the patents in the collection which we have marked for identification. In fact, it carries the tab number eight in the indexing. Now, may I hand this to the witness?

The COURT. Yes.

By Mr. GRAHAM:

Q. I hand you this index. The patent which I have identified, number 2,524,218, is the important one on which our continuity depends. And I will ask you now to refer to column one of the patent, right on the first page, at lines 25 through 46. And ask you to read that portion.

A. * * * All right.

Q. At this point I will ask the clerk to mark this card carrying a reproduction of the formula in that section.

Now, does that paragraph you have just read give us the formula reproduced in this Exhibit three?

A. Yes; it does.

Q. Now, will you describe the contents of that paragraph of section ——

Mr. SCHIMMEL. May I interpose an objection? I think the paragraph is plain enough for all to see and to read. And unless your Honor feels definitely

that it needs explanation, I do not think that the testimony of the expert is relevant to this case.

The COURT. Well, I take it this testimony is being offered to show what the starting point of the continuity is, is that right?

Mr. GRAHAM. That is correct.

The COURT. And, of course, the Court should know more than it does just now about what that is. Those are just simply some figures to an ordinary layman. My chemistry instruction consisted of the course that you get in the high school, and that is all, one year of chemistry in high school.

I would want to know what this really is so that I can follow the rest of the testimony to see what the subsequent developments were.

Mr. SCHIMMEL. As I said, your Honor, if you feel it is necessary, to understand the case, it is perfectly all right. I am agreeable. I was raising the objection just for the record.

The COURT. Very well, the objection will be overruled.

By Mr. GRAHAM:

Q. Now, Doctor Martell, does that paragraph when read with the formula, give you one formula or does it give you several formulas?

A. It gives several formulas.

Q. I will ask the clerk to mark our next exhibit for identification. Is one of the formulas given by that paragraph the formula I hold in exhibit No. 4, which has also been posed on the board?

A. It is.

Q. At this time I would like to introduce a third card marked for identification number five. Is the formula on this card, which has been numbered as five, included within the group of formulas in the paragraph we have inspected?

A. Yes; it is.

Q. Now, will you explain how that paragraph, which includes these formulas here, gives you that conclusion, and for this purpose, I will provide you with pencil and paper.

A. One takes the general formula given on exhibit three and applies the definitions in the following paragraph of the patent to the general terms given in the formula.

Now, if we take n as 2, it states here n is equal to two or three. If you take n as equal to two——

Q. That is the small subscript here?

A. The small n, that gives us two carbon atoms in a row as indicated there. m may be zero or a positive integer. Let us select zero.

Q. Will you explain why you select zero?

A. Because that would be a logical approach as one selects the simplest formula. And that drops out the group in parenthesis.

Q. Does that then bring us to the formula we have in exhibit number four?

A. Yes; that takes care of m and n, leaving only A and D to be defined.

Now, D is a member of the group consisting of acetic acid and propionic acid giving $-CH_2COOH$ or $-CH_2CH_2COOH$. So, acetic acid would give the two $CH_2OOOH$ groups at the bottom on the formula at the right.

Finally, A is defined as consisting of one of a number of groups, and if we let A equal to a hydrocarbon group having no more than two—or rather having at least two carbon atoms.

Q. Why do you select two carbon atoms?

A. Well, it states here at least two carbon atoms. One would usually start with the simplest structure.

And having not more than two hydrogens replaced

by one of a number of groups. If we replace one hydrogen by OR, which is allowed here in the list of groups——

Q. Where do you find that OR in the text of the patent?

A. At line 43, column one OR is one of the groups listed. Where R is alkyl or hydrogen and if R is hydrogen then we obtain $-CH_2CH_2OH$ for A.

Q. Why do you make R hydrogen?

A. If R is hydrogen; yes.

Q. Why do you make it hydrogen?

A. Because there again it is the simplest possible structure, and most chemists start with the simplest compound.

Q. Would it also be because hydrogen is specifically named at line 44?

A. It says alkyl or hydrogen. Alkyl may be any number of a group and hydrogen is a specific group; yes.

Q. Now, going back to the reason why you dropped out the parenthetical expression here, you mentioned you made m equal zero because it is logical. Would it also be because m is specifically defined as being zero in the passage we have quoted?

A. Yes; it was defined here as zero.

Q. Would it be fair then to say that that formula given at line 30 in the passage we are looking at, plus the following explanatory text, then dictates this specific formula given in exhibit five?

A. It does.

Q. I would like to have those exhibits marked and entered in evidence.

Mr. SCHIMMEL. No objection.

The COURT. They would be duplicates of the smaller cards that I hold?

Mr. GRAHAM. Yes. We had these reproduced on a larger scale.

The COURT. Very well, received in evidence.

(Plaintiff's exhibits 1 through 5 were marked for identification and received in evidence.)

Mr. GRAHAM. Now, I have the certified record of the Patent Office of case 2,524,218, and ask that that be marked for identification. This is a copy of the proceedings taken in the Patent Office.

The COURT. Regarding the application now in dispute?

Mr. GRAHAM. No; this was the application which matured into the patent we have been inspecting. This is the one marked Case A. I now turn to page 13 of that record, and may I offer it to the witness?

The COURT. Yes; that will be received.

(Plaintiff's Exhibit six was marked for identification and received in evidence.)

By Mr. GRAHAM:

Q. Will you read claim one, which is on page 13 of that certified record?

A. * * * All right.

Q. Now, we note that claim was initially filed in this application which matured into the patent which is case A, and the one which starts this chain of continuity. Does that claim give us the formula for the compound in exhibit three?

A. It does.

Q. Does it give us the formula for the compound in exhibit four?

A. Yes.

Q. Does it give us the formula for the compound in exhibit five?

A. It does.

Q. You note that the compound, that was an initial claim in the application, it specifically identifies the

compound for the Court, and it is part of the initial disclosure made to the Patent Office.

Mr. GRAHAM. We will now ask that that file record be entered in evidence as our exhibit.

The COURT. That has been received as exhibit six.

Mr. GRAHAM. Now, we turn to our chart of continuity, to what we have identified as Case B, and this is one of the applications which had to be continued out of the originally filed one. I have here the certified copy of the record of proceedings in the Patent Office involving Case B, which was serial number 39,377, and ask it be marked for identification?

(The document referred to was marked for identification as "Plaintiff's Exhibit No. 7.")

By Mr. GRAHAM:

Now on page 11 is the page carrying the claims which were filed in that case B. Will you note claim one as filed in that application?

A. On page 10?

Q. Page 11.

A. Yes.

Q. Now, I ask—I have another formula reproduced on a card, and ask that it be marked for identification.

(The formula referred to was marked as "Plaintiff's Exhibit No. 8" for identification.)

Mr. GRAHAM. Your Honor, that application was abandoned and will not be included in the collection.

By Mr. GRAHAM:

Q. Do you find that formula reproduced in the claim you have inspected, claim one of Case B, as filed?

A. I do.

Mr. GRAHAM. I have another formula reproduced on the card which I ask to be marked for identification.

(The formula referred to was marked as "Plaintiff's Exhibit No. 9" for identification.)

By Mr. GRAHAM:

Q. Do you find this formula included within the formula you are inspecting, or case B?

A. I do.

Mr. GRAHAM. I have another formula reproduced on a card which I will ask to be marked for identification.

(The formula referred to was marked as "Plaintiff's Exhibit No. 10" for identification.)

By Mr. GRAHAM:

Q. Is this formula which we have marked Exhibit No. 10, given by the claim you are inspecting?

A. Yes; it is.

Q. In other words, the claim you are inspecting is written in this form, but reducable to the form in exhibit 9, and reducable to the form in exhibit 10?

A. Yes.

Q. I ask you now to read claim five of Case B as it was initially filed. That is a specific claim included in the application. And will you, using the pad of paper you have before you, write out the formula for diethanol ethylene diamine diacetic acid.

A. * * * All right.

Q. Now, is that the same formula that we have in Exhibit 10?

A. It is.

Q. In other words, that is the same formula we have here in Exhibit 10?

A. Yes.

Q. It is the same we have here in exhibit No. 5?

A. Yes.

Q. Directly dictated by the claim?

A. Yes.

Q. I ask that this formula be marked as Exhibit and entered in evidence.

The COURT. Very well.

(The document referred to was marked "Plaintiff's Exhibit 11," and received in evidence.)

Mr. GRAHAM. Now, we come to the case which has been labeled in our chart as Case No. C, and that is the specific application which is in dispute with the Patent Office.

I have here the certified copy of the file wrapper and contents which I ask be marked for identification.

(The document referred to was marked "Plaintiff's Exhibit No. 12" for identification.)

Mr. GRAHAM. This application was filed on March 13th, 1951, as a direct substitute for Clase B, which was abandoned immediately after the filing of this one, and the chart directly shows the overlapping in time sequence of these, so I don't think we need note further these are copending cases, and the material was continuously before the Patent Office.

Now, may I approach the witness?

The COURT. You may. I was looking at those dates, and I wanted to get that in my mind. I see that the application in dispute was filed March 13, 1951, and the other was abandoned March 21st, 1951.

Mr. GRAHAM. There is material in the file which expressly identifies this as the continuation of the one which was abandoned.

In other words, the sequence of filing abandonment was a deliberate one, in which the application case B was not abandoned until there was in existence a filing receipt for the one before the Court.

The COURT. I see.

Mr. SCHIMMEL. May I say that there is no dispute as far as we are concerned as to Case B. We concede that Case B is a proper continuation of the presently

involved case and that it affords a basis for the claims in suit. We concede that.

Mr. GRAHAM. I think you have your identification of cases—Case B is not a continuation of C. C is a continuation of B.

Mr. SCHIMMEL. C is a continuation of B. We concede you have a basis in case B for the claims in suit. And that it was copending.

Mr. GRAHAM. Now, I have here reproduced, another formula taken from Case C, mounted on a card, which I would like marked for identification.

(The document referred to was marked "Plaintiff's Exhibit No. 13" for identification.)

By Mr. GRAHAM:

Q. Now, turning to the application of Case C, which we have before us, the certified proceedings, will you inspect page one of that application and tell us, Doctor Martell, does it state this formula which we have in Exhibit 13?

A. It does.

Mr. GRAHAM. At this point I would like to introduce another card containing another formula from Case C, marked for identification——

The CLERK. Number Fourteen.

Mr. GRAHAM. Number fourteen.

(The document referred to was marked "Plaintiff's Exhibit 14" for identification.)

Mr. GRAHAM. Now, I will ask the witness to turn to Claim two of the application as was filed. I am a little more familiar with the record, so I will ask your Honor if I may turn to it for him?

The COURT. Yes; you may.

Mr. GRAHAM. It is page 10 of the certified record. Claim two is the elected species within the Patent Office.

In other words, this application was filed, and as always happens in chemical cases, the Patent Office at first refused the broader claim and requires an election of a specific one for argument before them. This was the one which was elected.

By Mr. GRAHAM:

Q. Now, Doctor Martell, does that claim two give us the formula which we have here in Exhibit 14?

A. It does.

Mr. GRAHAM. Now, I would like to introduce another card carrying another formula, marked for identification——

The CLERK. Fifteen.

Mr. GRAHAM. Fifteen.

(The document referred to was marked "Plaintiff's Exhibit 15" for identification.)

By Mr. GRAHAM:

Q. Does Claim two of this application, Case C, specifically give us the formula for the compound which we have in Exhibit 15?

A. It does.

Mr. GRAHAM. I ask now that the file wrapper be received in evidence together with all the other exhibits we have introduced.

The COURT. They will be received.

(Plaintiff's Exhibits 7 through 15 inclusive, previously marked for identification, were received in evidence.)

Mr. GRAHAM. Now, this progressive filing of continuing applications was the result of Patent Office requirements, because the examining of patent applications goes according to Patent Office classifications. And when an individual comes in with a concept which is a little too large for Patent Office Classifications, he usually is faced with requirements for divisions.

These requirements are sometimes rather difficult to take, because they result in the filing of multiple applications. And in this situation we invite attention to the fact that in our Case A, Doctor Bersworth presented a broad concept of this type of compound. This was in his initial disclosure of this term.

He was very quickly faced with a requirement of division, and to complete this picture showing what happened to the claims in the case, another application was filed, which had not been marked as part of our chain of continuity, but it is this top one on the chart. It is U. S. Patent, which resulted in U. S. Patent 2,532,391.

Now, the certified record of U. S. Patent 2,524,218, will show that as initially filed, the application contained claims to compounds as such written in the form we have in Exhibit No. 3. And to detergent compositions constituting blends of detergent materials with compounds of this type.

Earlier in the prosecution of the case, the requirement of division was received and a divisional application was filed reasonably promptly afterwards.

That divisional application, or continuation, as it was called, continuation in part, has resulted in issue of U. S. Patent 2,532,391. It was filed March 15th, 1947, and I ask that the certified file wrapper be marked for identification.

(The document referred to was marked "Plaintiff's Exhibit 16" for identification.)

Mr. Graham. Now, in the certified wrapper of this U. S. Patent 2,532,391, we merely call attention to the claims as they were filed, because it speaks itself as a duplicate of that in our case A.

By Mr. Graham:

Q. I ask the witness to read claim one to himself.

A. All right.

Q. Does that claim give us the formula of the compounds which are given in Exhibit No. 15?

A. Yes; it does.

Q. Does it give us the formula for the compounds included in Exhibit No. 4?

A. Yes.

Q. Does it give us specifically the compound in Exhibit No. 5?

A. Yes; it does.

Q. All right. Let's take this next one. Well, these are duplicate formulas. And so are these, and so on, but does it give us the formula for this compound which is Exhibit No. 8?

A. Yes; it does.

Q. Does it give us the formula for the compound shown in Exhibit No. 9?

A. Yes.

Q. Does it give us the formula for the compound shown in Exhibit No. 10?

A. Yes.

Q. Does it give us the formula for the compound shown in Exhibit 13?

A. Yes.

Q. Does it give us the formula for the compound shown in Exhibit No. 14?

A. Yes.

Q. Does it give us the formula for the compound shown in Exhibit No. 15?

A. Yes.

Mr. GRAHAM. I ask that it be noted this case was filed March 15, 1947. This is the initial claim in the case also, and can be considered disclosure as such. It is in the same form as the disclosure in U. S. Patent 2,524,218.

Now, that is the necessary story of continuity on this subject matter, and our efforts to claim it before the Patent Office.

Now, we can refer briefly to the point where the initial disclosure was made. And at this stage we turn to our collection of references, and I will ask the witness to turn to U. S. Patent 2,428,353, Index tag No. 5 in the collection.

By Mr. GRAHAM:

Q. Now, at page one, column two of the patent, lines 40 to 55, I ask you to inspect the generic formula there given.

A. * * * All right.

Q. Does that section of the disclosure give us a formula for compound in the form of Exhibit 4?

A. It does, yes.

Q. Does it give us a formula for the compound in the form of Exhibit 9?

A. Yes.

Q. Does it give us a formula of a compound in the form of Exhibit 14?

A. Yes.

Mr. GRAHAM. That is, your Honor, the initial disclosure which was made of this subject matter. It could not have been claimed in this application, which issued as U. S. Patent 2,428,353, because in that case compounds which are esters of these were being claimed. They have quite a different utility and constitute a rather different class of subject matter.

Now, we ask the witness to refer to the same patent now at column 7, over on the final page, lines 35 through 55, and invite attention there to the commentary on organic chemical nomenclature.

The COURT. Will you give me that reference again?

Mr. GRAHAM. It is column 7, lines 35 to 55, the final paragraph of the specification.

By Mr. GRAHAM:

Q. Now, is that an accurate commentary on organic chemical nomenclature?

A. Yes; it is.

Mr. GRAHAM. I am inviting attention of the Court to that passage purely for the purpose of showing that not only are these organic names long, but you can have several names for a given structure. And for convenience, chemists adopt the habit of talking in terms of formulas when they are talking of a more complicated compound, as witnessed by this name I wrote on the board. A specification of a patent which used names only would run on to many many pages if the names were all written out.

By use of formulas, in a few paragraphs of description, it is possible to condense a great deal of information into a single paragraph, both in the general and in the specific sense.

By Mr. GRAHAM:

Q. Now, one final reference in this collection of patents we are inspecting, which is U. S. Patent 2,413,856, and it is tagged No. 4. And we invite the attention of the witness to column one, lines 39 through 50. That is the generic—or, you describe that.

A. This gives a general formula which is an alkylene diamine diacetic acid with Y and Y, with a little dash beside it.

Q. Now, does that passage give you a formula for compound in the form we have in Exhibit No. 4?

A. Yes; it does.

Q. Does it give you a formula of a compound in the form of Exhibit 9?

A. It does.

Q. Does it give you a formula of the compound we have in Exhibit 14?

A. Yes.

Mr. GRAHAM. Another difficulty which arose after we had shown continuity in the Patent Office was a certain German patent which was issued back in 1937.

Now, usually, in a case of this kind, arguments in reaching back for early disclosure are made for the purpose of getting behind an anticipation in an adversely held reference.

We can't get back, or even begin to get back to this German reference. In other words, 1937 was some six years before we were in the Patent Office with our earliest disclosure. And are pointing to our own earliest disclosure to indicate when the basic concept of these compounds had been conceived.

It was presented to the Patent Office and thereafter diligent effort was made to have the claims accepted.

Having established continuity at all, continuity before the Patent Office, we were told at one stage by them that if we read our U. S. Patent 2,524,218 to find these specific disclosures we have been demonstrating in Exhibit 3, 4, and 5, then the Patent Office says they are entitled to read the broad disclosure appearing in this German Patent 642,244, to find in it an anticipation of the compound we are claiming. And that will be the purpose of the discussion of this German Patent with the witness.

We ask the witness now, to turn to this German patent which is tab No. 7 in the collection, and since he reads German fluently, he won't have to use the translation which has been provided, but there is a translation which has been keyed to the text, by marking corresponding lines.

Now, referring to that German patent, Doctor Martell, will you inspect the passage on page one, commencing at line 30, and running through about line 37?

The WITNESS. Yes, sir.

By Mr. GRAHAM:

Q. Will you describe what that passage tells you?

A. It states that the author of the patent has found that certain amino compounds and amino acids are effective in the washing and purification of gases.

Q. In other words, he is describing a process for separating gas mixtures by contacting the gas mixtures with an amino compound?—

A. That's right.

Q. For purposes of contrast, that is the basic theory of the invention described in this case. Now, we ask you to turn to page two of the German Patent, referring to lines 29 through 75.

A. * * * I have done so.

Q. Will you describe what this passage includes?

A. Well, four general formulas are presented, each containing one nitrogen, and a number of groups are attached to the nitrogen atom and to other chelates, and these groups can be any number of various compounds, some of which are acetic, some of which are hydrocarbons and some are hydrogen, others are alkyloxyl, and so on. This is a group of general formulas.

Q. All right. In other words, it is a group of general formulas, four to be specific, plus explanatory text, telling you what values to put in to those general formulas to derive compounds that will be useful in the processes described in the patent?

A. Yes.

The COURT. Let me interrupt. Why do you discuss the German Patent? What is your logical connection there?

Mr. GRAHAM. The logical connection is this: It came out of the action of the Board of Appeals in the Patent Office. They, in their action, which Mr.

Schimmel had reference to, acknowledged that our continuity depends on our own U. S. Patent 2,524,218. They even acknowledge that patent comes up with this specific compound, but they go on to say that if we are entitled to read that patent to find that specific compound, then the same applies to them. They can read this German patent, which we thought we had buried in the Examining Division, can be read the same way to derive the same compound.

The COURT. They are citing now the German patent as anticipatory?

Mr. GRAHAM. After we disposed of it in the Examining Division. And, as I say, for complete presentation, we must present our views on the significance of it.

Mr. SCHIMMEL. May I make a statement in that connection, your Honor?

The COURT. I would be glad to hear from counsel.

Mr. SCHIMMEL. I think the crux of this case depends on the holding of whether or not the Plaintiff is entitled to go back to the case on which he is asserting his right for the specific claims in suit here. I would say this: If he convinces your Honor that he is entitled to rely on the general formula disclosed in the Case A, I don't think we need worry about the German patent. I am willing to eliminate that issue from this case. I think we should tie it down to the one proposition, as to whether or not he is entitled to rely on Case A for the claim of subject matter. I am willing to drop any other contributions.

Mr. GRAHAM. Well, we have labored carefully to demonstrate how that general disclosure in our Case A says this, and this, and this [indicating] and mainly, what we have in formulas 3, 4, and 5. And we will not discuss the German patent in detail.

The COURT. That seems clear to me and proper under the circumstances. I think we might take a

little rest here. This is a very great strain on the reporter.

Mr. GRAHAM. That also completes this witness' testimony.

The COURT. Do you wish to cross-examine?

Mr. SCHIMMEL. Oh, yes; certainly.

The COURT. Very well. We will take the usual morning recess then.

(Whereupon, a short recess was taken.)

The COURT. You may proceed.

Mr. SCHIMMEL. I believe that it might be apparent to your Honor that the general formulation that has been testified to by the witness is the same in all three cases.

Cross-examination by Mr. SCHIMMEL:

Q. But I think, in order to make it positive and definite, I would like to ask the witness if that is his opinion, that in Case A, Case B, and Case C, as you remember them, is the general formulation identical?

A. Yes.

Q. Now, let's turn to the Patent 2,524,218, in the folder, which is item 8 in this folder that Plaintiff's counsel presented. Looking at the general formula in lines 25 to 45, the explanatory language in connection with the letters used in that formula, can you give us any idea of how many compounds are encompassed within the scope of that general formula?

A. I would say a large number.

Q. Would it be unreasonable to say that it might run into the thousands?

A. I would say practically, no, but possibly; yes.

Q. From the theoretical aspect then, using the definitions given in the explanatory portion of the reference, you would say it would run into the thousands, is that correct?

A. When one sees—may I use my own words?

Q. Yes; go ahead.

A. It says here m is zero or positive integer. Therefore, one could conceivably say that m could be almost any number. Chemically speaking we wouldn't go to very large values of m, because compounds would be impossible to prepare.

Q. From the theoretical aspect, isn't it true that would cover many known as well as unknown compounds?

A. A small number of known, but many unknown.

Q. Many unknowns? Can you find in that designation of the general formula an express disclosure of the diethanol ethylene diamine diacetic acid compound which is the compound of the claim set forth in this case, and which is on exhibit to the right on all three columns, Exhibits 5, 10, and 15?

A. By inserting the allowed values of n, m, A, and D, the formulas 5, 10, and 15 are expressed; yes.

Q. I don't believe you understood. I don't believe you understood my question. Is the compound expressly disclosed as such?

A. I am afraid that I don't understand your meaning.

Q. Is the compound named, is it named?

A. The compound specifically is not named as such in this paragraph.

Q. You derive the compound, I gather, by assigning certain values to the m variables set forth in the general formula, is that correct?

A. Yes.

Q. But there are many other compounds that could be derived in the same way, isn't that correct?

A. Yes.

Q. Let me direct your attention to Patent 2,413,856, which is item four in that same folder. Let's change that, item five, Patent 2,428,353. And directing your

attention to column 3, line 26, the compounds being named is ethylene, diamine, N, N', diacetic acid, N' ethanol. Is that compound one which falls within the scope of the generic expressions which were being discussed previously?

Mr. GRAHAM. May I interpose an objection, your Honor. That is not the compound we are arguing about. But it is the compound defined in the non-elected portion of the application before the Court.

It is instructive to inspect that compound, however, to recognize that it is in the same application. However, it is not the one on which we specifically presented our evidence, because that is not the elected claim.

The COURT. In view of that statement, do you still wish to press your question?

Mr. SCHIMMEL. Yes, Your Honor, because I want to show certain matters were abandoned by the applicant by virtue of these specific things, and that is part of our case. I concede it is not the express and specific compound before your Honor. But it is a compound that is encompassed within the broad expression which counsel for Plaintiff had the witness testify to. And to that extent, I think it is quite pertinent to this case.

The COURT. He may answer then. The pertinence of it may be argued later.

By Mr. SCHIMMEL:

Q. You may answer.

A. I would have to look again at the general formula and explanatory paragraph to apply this particular formula.

Q. Suppose you do that.

A. Which one?

Q. The one we were speaking about previously, that one.

A. The general formula indicates two A groups, which are alike.

Q. Would you mind pointing out wherein it says they have to be alike?

A. A is a hydrocarbon group, containing at least two carbon atoms and having not more than two hydrogens replaced by a member of—and then the OR comes in at that point.

Q. Is there anything that says definitely they have to be the same?

A. Well, first it defines D as being diacetic acid. And then it defines A as a hydrocarbon group, containing at least two carbon atoms and having not more than two hydrogens replaced by a member of the series—of one of the series as an OR group where R is hydrogen giving ethanol.

The statement that A is a hydrocarbon group, I would say infers a single hydrocarbon group. However, perhaps the Patent Office has some other interpretation of that.

Q. But do you find in there anything that says A has to be identical on each nitrogen atom?

A. It is not specifically stated.

Q. Would you normally interpret that to mean it would have to be identical under the broad definition given there?

A. When I see a general formula showing two A groups alike, I usually, normally come to that conclusion.

Q. But wouldn't it be true that under the general formulation you could have an ethyl group as one of the A's and propyl group on the other A?

A. I think the inference is the other way around, however, it is not definitely stated one way or the other.

Q. Would it be a violation of that general formula to say that a compound that had the ethyl group on one A and a propyl group on the other A would not come within the scope of that general formula?

A. Normally, I would not read the general formula to give different A groups.

Q. So then, do I gather from your answer that you would say that ethylene, diamine triacetic acid ethanol would not be within the scope of that general expression?

A. Triacetic acid ethanol? Would you repeat the question?

Q. The compound named on column 3, line 26, of Patent 2,428,353.

Mr. GRAHAM. May I object at this point? Again, I wish to remind people that Mr. Schimmel is questioning the witness about the nonelected subject matter of the application we have before the Court. In fact, that subject matter is in a divisional application in the Patent Office, in which we are arguing the patentability of that compound. And we have stipulated with Mr.—the solicitor from the Patent Office, that the claims of the application before the Court, which read on this triacetic acid monoethanol compound may be dismissed from this suit without prejudice. I have the stipulation. We plan to hand it up at the termination of the presentation of evidence. And that is the specific reason we confined our showing to the diethanol compound which are these [indicating].

Mr. SCHIMMEL. May I say this, your Honor: Not withstanding that we have agreed to limit this case

to the consideration of the diethanol compound, the witness has testified to something, as I understand his testimony anyway, which is contrary to what the inventor himself said.

The inventor in this application has said that the mono-substituted compound is within the scope of the broad generalization.

Mr. GRAHAM. May I comment on that? That came up immediately in the Patent Office after the filing of the application now before the Court. And the first thing the Patent Office told the inventor was to elect his species and confine his argument to that elected species. During the history of the application before the Patent Office, argument was made on this elected species, namely this compound [indicating]. But they took gratuitous slaps at the monoethanol compound, rejecting it as nonelected subject matter and for other reasons. That was the other reason we have specific complication with that German reference, which Mr. Schimmel has chosen to leave buried in the Patent Office.

The COURT. The difficulty the Court has is to rule on these questions of evidence and objections to evidence before I have come to a thorough understanding of what the issue is.

I think it would be best at this time to let the witness answer the questions, and then later, in your brief or in your argument you may show your authority for objection.

Mr. GRAHAM. I will be perfectly happy to preserve the objection.

The COURT. Very well.

Mr. SCHIMMEL. What was your last answer?

Mr. GRAHAM. I don't believe the witness answered any last question.

The WITNESS. I think I answered it.

The COURT. That's right.

Mr. SCHIMMEL. Then let me get it clear. I asked the witness whether the compound was named, and I don't believe that was specifically answered by the witness.

The COURT. No; it was not.

By Mr. SCHIMMEL:

Q. I will ask you then does that compound named in line 26, come within the broad designation?

A. Of which broad designation? In this patent?

Q. Of that shown in Exhibit 3?

A. As I indicated before, normally I would not include that formula.

Q. I show you page 10 of the application involved in this suit, which I believe is Plaintiff's Exhibit 12, page 10. Directing your attention to claim one, would the compound in line 26 of the patent 2,428,353 that we were referring to previously, come within the scope of that claim?

A. Would that compound come within the scope of what?

Q. Claim one of the present application which appears on page 10 of Plaintiff's Exhibit 12?

Mr. GRAHAM. May I interpose another objection at this point? I wish to remind people again that the solicitor for the Patent Office is talking about the nonelected subject matter, which is in the divisional application in the Patent Office, properly a divided case, based on requirements of the division in the history of this application. And in as much as he chooses to stay on the nonelected subject matter I would like to preserve a continuing objection on this entire line of questioning.

The COURT. Yes; certainly. The record will show that a continuing objection exists.

By Mr. SCHIMMEL:

Q. Will you answer?

A. The answer is that Claim one does not. I would not normally conclude it as conforming to the general formulas shown by Exhibits 3, 8, and 13. And it does not conform to the Exhibits 4, 9, and 14.

Q. That wasn't the question.

A. I'm sorry.

Q. Looking again at the Patent 2,428,353, which is number five in the folder, and considering the compound named in line 26, column three, the ethylene diamine triacetic acid ethanol——

A. Yes.

Q. The question is, does that compound come within the designation, broad designation, of claim one of the presently involved case?

A. Yes.

Q. You are familiar, I gather, with every case referred to. Do you recollect whether that same compound, the ethylene diamine triacetic acid ethanol, was disclosed in the case that is referred to as Case A in this proceeding, which is Patent 2,524,218? A certified copy of that application which resulted in that patent, I believe is Plaintiff's Exhibit 6.

A. Now, the question is whether ethylene diamine triacetic acid monoethanol is disclosed——

Q. Specifically in that patent?

The COURT. The German patent?

Mr. SCHIMMEL. No; in Bersworth's Patent 2,524,-218, which is in the folder.

The WITNESS. I would have to have time to read the whole patent, but on a quick inspection I note that all the compounds in this patent seem to be symmetrical, which is not the case with the triacetic acid. I would conclude it is probably not here.

By Mr. SCHIMMEL:

Q. Now, in connection with that same patent, 2,524,218, I believe you answered a question asked of you as to whether it was fair to say that the formula, general formula, given in that patent dictates the specific compound claimed in this case, is that correct?

A. Yes.

Q. When you say it dictates that specific formula, do you mean that specific compound is named specifically?

A. No.

Q. Do you mean that it is named inherently?

A. I mean that the formula leads directly to the compounds.

Q. Now, isn't that formula, as you have previously stated, one which names, under your terminology, I believe, thousands of compounds?

A. Theoretically; yes.

Q. Would you say that such a broad general indefinite designation names anything specifically?

Mr. GRAHAM. I would like to object to that question. He has characterized this disclosure as a "broad, general, indefinite one."

Mr. SCHIMMEL. I will remove the "indefinite" from the question.

Mr. GRAHAM. Will you repeat the question now?

Mr. SCHIMMEL. Would you say the designation of that broad general formula encompassing thousands of compounds, names any specific compound?

The WITNESS. I would say it leads directly to a specific compound.

By Mr. SCHIMMEL:

Q. But would you say it names the compound?

A. It names a class of compounds, not any specific compound.

Q. Therefore, when you say that the formula dictates the compound shown in the exhibits on the right hand side on the blackboard, that is, the specific diethonal compound——

A. Is that the question?

Mr. SCHIMMEL. Will the reporter read the question?

(The reporter read the question as follows:

"Question. Therefore, when you say that the formula dictates the compound shown in the exhibits on the right hand side on the blackboard, that is, the specific diethanol compound——").

Mr. SCHIMMEL. You do not mean that it names the compound, is that correct?

The WITNESS. It names a class of compounds and leads a chemist directly to the compound on the right, the exhibit there.

By Mr. SCHIMMEL:

Q. Just as it would lead a chemist directly to thousands of other compounds, isn't that correct?

A. Not the same way; much more directly.

Q. But, it would be fair to say it would lead him to a great number of compounds in the direct same path that you used to get that specific compound?

A. No; the path I would take would be to look for the simplest combination of groups.

Q. Wouldn't the specific designation also give you the propionic acid compound?

A. Not as logically. One would look for the acetic acid first.

Q. Isn't the propionic acid specifically named?

A. Yes; that's right.

Q. And, then wouldn't it be just as logical and reasonable to get the propionic compounds as well as the acetic acid compounds?

A. If I were going to prepare the substance, I would choose the acetic acid first.

Q. Looking at Patent 2,524,218 again, I believe you stated that the diethanol compound was not specifically named in that patent, is that correct?

A. Was the question, was diethanol specifically named in the general formula?

Q. No; just whether the diethanol ethylene diamine diacetic acid was specifically named?

A. In the whole patent?

Q. In the whole patent?

A. I would have to read the patent to be sure of that, sir; I would have to read it.

Q. In other words, you do not feel you are familiar enough with the patent or the case to answer that question?

Mr. GRAHAM. I object to that question. The witness is completely familiar with the patent. And I will say for the record that the name of diethanol ethylene diamine diacetic acid compound has not been spelled out in it.

Mr. SCHIMMEL. Would you also, Mr. Graham, make the same admission with respect to Patent 2,413,856?

Mr. GRAHAM. I would have to review that one more closely. I don't believe it is named there.

Mr. SCHIMMEL. Would you make the same admission with respect to 2,532,391, which you said was identical, I believe, you said?

Mr. GRAHAM. The name of the compound has not been spelled out.

By Mr. SCHIMMEL:

Q. Now, looking at application 39377, which is Case B on this chart, and which is Plaintiff's Exhibit No. 7, I believe you testified that Claim two of that application, which is on page 11, is directed to the diethanol ethylene diamine diacetic acids named,

which is shown structurally on the exhibits 5, 10, and 15.

A. No; I stated that Claim 5——

Q. Pardon me.

A. Named the compound.

Q. Claim 5 names the compound which is shown structurally on Exhibit 5, 10, and 15, the ones on the right hand side?

A. Yes.

Q. Is that correct?

A. Yes.

Q. From your knowledge of the applications in this case, would you be prepared to say as to whether or not that is the first time that specific compound was named in any of these applications involved?

A. There are so many applications that I can't specifically remember.

Mr. SCHIMMEL. Would you care to make a statement on that, Mr. Graham?

Mr. GRAHAM. We will inspect the record to determine whether that is true.

Mr. SCHIMMEL. I think that is all I have, Your Honor.

The COURT. Very well. Any redirect examination?

Mr. GRAHAM. I have a few questions.

Redirect examination by Mr. GRAHAM:

Q. I invite the witness' attention to the Patent 2,524,218 which is our Case A, and before asking him any questions, I want to comment on this paragraph in disclosure in column 1, lines 25 to 46.

That paragraph is a creature of the Patent Office practice. It seems that in claiming a chemical compound quite a number of years ago, an individual named Markush ran into some trouble trying to classify a group of compounds together for the purposes of a chemical claim. He tried the one way

and the Patent Office said no good; he tried the alternative way and they said no good; he tried it another way and they still didn't like it. He settled it by appeal to the Commissioner in which the Commissioner of Patents specifically approved language which says a compound selected from the group consisting of, and then listing the ones that he had.

In other words, the ruling in the case specifically was that where the English language is so poverty stricken as to restrict a class of compounds, you can add them up by identifying them separately.

Now, under that practice there has developed a large amount of literature within the Patent Office as to how you set up proper Markush claims.

And the proper way is to set up a formula the way we have it written at line 30 of this paragraph. Then you take the individual groups, these A and D, which we have in the formula, and define what they are. That is in your specifications. Then, in your claims, you draw a claim in that form.

We invited attention to this patent application as initially filed, and it contained a claim drawn in the Markush form, incorporating essentially the disclosure from lines 25 to 46. The said compound corresponding to the formula given at line 30, wherein n is two or three, and so forth. Specifically using the language as it was set forth here. That form of claim has carried over into each of our continuing cases.

What that language means to chemists is not one formula, it means a formula for each member of the group as specifically included in that kind of language. And if we can take a few minutes we can demonstrate that on the blackboard. It has been done in the record once for the Board of Appeals, when we were requesting consideration of this case. But

we can do it briefly on the grease board without complicating matters too much. I hoped to avoid this kind of demonstration——

Mr. SCHIMMEL. This is actually a question of argument, isn't it, rather than evidence?

Mr. GRAHAM. We have taken evidence on the point. There are specific questions in which Doctor Martell testified that it is not one formula at that point, but it is a group of formulas.

Mr. SCHIMMEL. Also thousands.

Mr. GRAHAM. Of the group, one of them is specifically the compound we are claiming, and I will correct Mr. Schimmel; it is not thousands. Infinite, it could be a group of compounds, but the application is self-limiting; the subject matter here is restricted to water soluble compound. Compound that will go in the solution like the solution your Honor has on the bench before you, and that automatically limits this disclosure to quite a limited class of compounds.

Mr. SCHIMMEL. Mr. Graham seems to be testifying.

Mr. GRAHAM. I have lived with the case long enough to feel expert enough to do so.

Mr. SCHIMMEL. I shall direct your attention to the Claim. It says nothing about water solubles. The claim is broad and unlimited. And that particular compound, and the generic expression was any compound coming within the broad generalization of it. The witness has conceded that it covers innumerable unknown compounds as well as some known compounds, which might run into thousands. So all we are doing is arguing the case at this point, it seems to me, and I think that should be reserved until later.

That is the basis, as the decision developed below, the basis is, does a generic broad expression entitle one to claim a specific thing. We say the law says

no, you can not do that. That is what we are arguing about.

Mr. GRAHAM. If the disclosure specifically brings you to a specific compound, you are entitled to claim it. And I can demonstrate where we have done that.

The COURT. There is nothing for me to rule on now. Counsel already said he was making a statement, and I am glad to hear counsel, because I wish to be kept on the beam, so to speak. And I want to know what your contention is regarding these bits of evidence that you are offering. And I welcome the statement and will not take it as evidence. It is not offered as such. Merely calling the Court's attention to things that are in the record, what your contention is.

So, do you have questions now? I am wondering if it is necessary to go as far as you have indicated you wish to go at this time, in view of the exchange that we have had here. It becomes a question of law and of argument.

Mr. GRAHAM. It is a matter of disclosure and I can do it in about three questions.

The COURT. Very well.

By Mr. GRAHAM:

Q. Referring now to this disclosure, Doctor Martell, at line 30, there is a complicated formula given in our Exhibit No. 3, in Patent No. 2,524,218. We went directly from the formula in Exhibit 3, through the formula at 4, to arrive at this one and find the specific disclosure of the compound we are claiming.

Now, coming back to Exhibit 4, could we have put in A positions, the first substituent groups suggested in line 42?

A. Yes.

Q. It would have been equally specific as this?

A. Yes.

**Q.** Could we have put in the A positions the second substituent in line 42?

**A.** Yes.

**Q.** Could we have put in those positions that first substituent that appears in line 43?

**A.** Yes.

**Q.** That would be a sulphonate compound. In that way could we have gone through that list of specific substituents?

**A.** Yes.

**Q.** The next, to last step would have been the OR?

A. That's right.

**Q.** Which is the compound we are claiming, is that correct?

**A.** That's right.

**Q.** That is the specific disclosure; those are the steps that lead directly to the compound we are claiming.

For a few minutes, Mr. Schimmel was on the non-elected species, and made a point of the fact that it is not named in our Case A. I will invite the attention of the witness to column one, line two. Do you find there, reference to U. S. Patent 2,407,645?

A. I do.

Mr. GRAHAM. I will offer the certified copy of the wrapper and contents of that application, 2,407,645, which is number seventeen.

That is incorporated by reference right into our Case A. And now, I invite the attention of the witness to page 8 of that certified wrapper and contents, and will you identify the fourth compound listed by formula?

The WITNESS. The monoethanol, ethylene, diamine, triacetic acid trisodium salt.

Mr. GRAHAM. Does that correspond to the Claim one, which Mr. Schimmel was asking you about?

The WITNESS. Yes; it does.

Mr. GRAHAM. Thank you. That will be all.

(The witness left the stand.)

Mr. GRAHAM. I call at this time as my second witness, Doctor Seymore Lewin.

The COURT. I wonder whether it is worthwhile. We adjourn at 12:30.

Mr. GRAHAM. Well, since Mr. Schimmel has chosen to bury the German Patent we will shorten Doctor Lewin's testimony, but perhaps it would be better to take our recess now.

The COURT. Very well. We will adjourn until 2 o'clock.

(Whereupon, a luncheon recess was taken at 12:20 p. m.)

AFTERNOON SESSION

2:00 O'CLOCK P. M.

The COURT. You may call your next witness.

Mr. GRAHAM. We have one thing to ask first. Doctor Martell, who was our principal witness this morning, has been away from his classroom in Massachusetts all of last week, attending the Chemical Society Convention in Texas, and then we had anticipated having him go on yesterday.

He is anxious to get back, and Mr. Schimmel, I have been informed, has nothing further to ask him, and we do not.

The COURT. He may be excused.

Mr. GRAHAM. Will you please call Doctor Lewin?

Thereupon Doctor SEYMORE LEWIN was called as a witness by counsel for Plaintiff, and having been first duly sworn, was examined, and testified as follows:

Mr. GRAHAM. I would like to explain very briefly the purpose in calling Doctor Lewin as a witness.

We have demonstrated the continuity of subject matter in this application this morning. The problem also involves a question of whether this patent on which we depend has a sufficient disclosure to maintain that continuity. And the purpose of Doctor Lewin's testimony is to be directed to that point.

Now, I have deliberately refrained telling him anything about the application or the case. In fact, he has read only the significant portions of the patent on which we depend for continuity. And, of course, he is a very observant individual and I imagine he has learned something about the case this morning. But that will be the nature of his testimony, to demonstrate what a skilled chemist can see in this disclosure.

The COURT. Very well.

Direct examination by Mr. GRAHAM:

Q. What is your name, sir?

A. Seymour Z. Lewin.

Q. And your address?

A. My address is 72 Seaman Avenue, New York City.

Q. What is your occupation?

A. I am at present, Associate Professor at New York University, Chemistry Department.

Q. Will you describe, briefly, your educational background and experience, mentioning publications and things of that Nature?

A. I have my Bachelor's degree from CCNY, Masters and Doctor's degree from University of Michigan. I received my Master in 1942, Doctor in 1950. Between 1942 and 1950 I worked as chemist teaching fellow, as lecturer and research fellow at the University of Michigan.

In 1950 I came on a post-doctoral fellowship to New York University and served as instructor pro-

fessor and associate professor. My work at the University of Michigan has been principally in the field of structure and characterization of organic compound, and I have continued this work at New York University.

I have over fifteen publications in the chemical literature. I serve as a consultant to the New York State Board of Education, and construct examinations for them in chemistry.

I am a consultant for the New York book publishing firms, and Editor of the Van Nostrand Chemist Dictionary.

Q. Is it fair to ask at this point whether you are familiar with the rules of organic chemical nomenclature?

A. I am; yes.

Q. Are these rules of organic nomenclature familiar to chemists generally?

A. Yes.

Q. Are these rules of nomenclature generally available to chemists, in the event they don't happen to remember them specifically?

A. Yes, they are, in all the standard handbooks and all the standard text books.

Q. Purely for the record, and I will not introduce this handbook we brought with us, but this is a typical chemist handbook carrying a chapter—it is the 32nd edition, 1950 to 1951 handbook of Chemistry and Physics, which, starting at page 640, carries a transcription of these standard rules of nomenclature. In the event we need refer to them, that will be one point.

Now, you heard Doctor Martell's testimony this morning on Organic Chemical nomenclature, what it means to a chemist, and his equating of a name for a chemical compound with its formula. Do you agree with that testimony?

A. Yes.

Q. In other words, you agree that a definitive name derived in accordance with these rules of nomenclature give the formula of a compound?

A. Yes.

Q. And do you also agree that the formula for a compound written in structural form will provide a chemist with a name for that compound?

A. With one or more names that may be equivalent; yes.

Q. In other words, several definitive names can be derived from a single structural formula?

A. Yes.

The COURT. Let me ask a question that has nothing to do with this case. Are the formulas for foreign patents the same that they are for ours, in English, or are they different?

Mr. GRAHAM. Yes; they are the same, or very similar.

The COURT. Those symbols are like symbols of music. They are an international language. Now, these names that you are referring to, would they be similar?

Mr. GRAHAM. They are very similar, but let me explain——

The COURT. You needn't take the time——

Mr. GRAHAM. Just to explain that, let me say that here, for example, the N, C, and H in this formula represent nitrogen, carbon and hydrogen, specifically. They are international symbols regardless of the language. The formula will be written with those Latin letters.

Now, these letters we have put on are arbitrary alphabetical designations to give us a basis for the finding of more chemical symbols to go in those positions.

Mr. SCHIMMEL. May I add, your Honor, just to clarify, that the arbitrary symbols will vary. They are not uniform.

The COURT. Yes. I take it that they would.

Mr. GRAHAM. That is up to the taste of the individual.

The COURT. And they depend on the description in the application?

Mr. GRAHAM. Yes.

The COURT. Science and art have a universal language. That is what interests me. And I am also of a very firm opinion that the basic rules of law are the same in all civilized countries. And why we do not have an enforceable system of law for the world is the $64.00 question before our day and age.

I didn't mean to interrupt you, Mr. Graham. Please continue.

By Mr. GRAHAM:

Q. I will now ask the witness to turn to our selection of patents, and to the Patent 2,524,218, Tag No. 8 in this index, and that is Case A in our chart, showing continuity. Will you inspect the passage appearing at column one, lines 25 through 46 on that patent?

A. All right.

Q. Will you describe the contents of that?

A. This passage contains a structural formula and a set of directions for substituting the symbols in the formula by chemical symbols, which will lead to a number of chemical compounds.

Q. Is the formula the one shown in Exhibit No. 3?

A. Yes.

Q. A general one. In your opinion, does that general formula, with the subsequent explanatory matter specifically bring you to the compound we have shown in Exhibit No. 5?

A. Yes; it does.

Q. I will now ask you to demonstrate why you have reached that conclusion. You may use a pad and pencil if you wish.

A. Well, I think I can do it by referring to the charts on the board there. The first chart on the extreme left would be the starting compound, at least, the starting structure. And the directions say where n is either two or three, then we choose two as the first example. If n is equal to two, this makes two $CH_2$ groups—what you are pointing to now; yes. And then it says m is either zero or positive integer. Zero is the specific number given here, so if we choose zero, that whole part in brackets is eliminated and this gives us then what you have on the second chart.

Q. This is Exhibit No. 4.

A. Then, on the second chart we have the symbols A and D. It says D is a member of the group consisting of $CH_2COOH$, or the first one; if we choose the second one that gives us the lower part of the extreme right hand chart, $CH_2COOH$ on the two nitrogens.

It continues. Wherein A is hydrocarbon group containing at least two carbon atoms. So, on each of the nitrogens we have two $CH_2$ groups. And having not more than two hydrogens. And if it says not more than two, let us choose one as the simplest case. Replaced by a member of the group consisting of—and the fifth member of that group would be OR, so we now have the structures on the top, $CH_2CH_2OR$, and OR having replaced a hydrogen.

And the text continues. Wherein R is alkyl or hydrogen. Hydrogen is the specific one involved then. And this gives us exactly the formula indicated.

Q. That is exactly the formula of Exhibit 5?

A. Yes.

Q. Would you say that that formula, with the explanatory disclosure, specifically dictates the formula we have referred to in Exhibit 5?

A. Yes.

Mr. GRAHAM. At this point we might invite the attention of the Court to Claim two of the application in issue. That is the one we have been over this morning. In fact, Claim two of the application in issue as written in this formula [indicating] and that is Exhibit 14, which is the identical form which Doctor Lewin has just gone through. And we made the point that it goes to this specific compound. In other words, it would come up specifically with that compound.

The COURT. You say that Exhibit 4 and Exhibit 14 are exactly the same. Then D and B would seem to have no significance.

Mr. GRAHAM. That is the arbitrary choice of the individual who did the writing of the application. At the time he wrote case A and seemed to prefer D for that position, and sometime later he uses B, but the definition of D or B is the same.

The Court. I understand.

By Mr. GRAHAM:

Q. In your opinion then, Doctor Lewin, would any skilled chemist derive the same information from a reading of the paragraph we have identified in U. S. Patent 2,524,218, which you have demonstrated?

A. Yes.

Mr. GRAHAM. That is the essence of Doctor Lewin's testimony. We were prepared to discuss this German Patent, but since it has been cremated, that is the end of my questioning of this witness.

The COURT. You may cross-examine.

Mr. SCHIMMEL. I just have a few questions, your Honor.

Cross-examination by Mr. SCHIMMEL:

Q. In describing a manner in which you arrived at the ultimate specific compound depicted on Plain-

tiff's Exhibit 5, I believe you said you used the first indication in the general descriptive language for all the arbitrary symbols, is that correct?

A. I would say the first specific indication.

Q. That was true with respect to n, wasn't it?

A. Yes.

Q That was true with respect to m?

A. Yes.

Q. That was true with respect to D?

A. Yes.

Q. Was it true with respect to the other member?

A. The matter of R, you mean?

Q. Yes.

A. Well, the text reads, wherein R is alkyl or hydrogen. Alkyl is not a specific group; it could be any of an infinite number.

Q. What would be the first one that would come to one's mind?

A. Methyl.

Q. You didn't select that?

A. No; I chose the first one mentioned. Methyl is certainly covered here. One would be led to the methyl compound too.

Q. As the first one, you did not select it to arrive at the final formula, as you have done in all the other designations?

A. Well, the answer to the question is no. And, if I may add, I would say if the text had read on line 32, m is either a positive integer or zero, I would have chosen zero, as I did, because that is the only specific thing given.

Q. Isn't one a positive integer that could come to one's mind normally?

A. I don't deny they are both possibilities.

Q. Let's get back to the next substituent. There are a group of substituents indicated for replacement

of the hydrogen atom. Did you pick the first one there?

A. No.

Q. Is there any reason why you shouldn't have picked the first one, just as you did in the other instances?

A. Oh, no. I understand that I was to explain how one is lead to this compound.

Q. One is lead to other compounds as well?

A. Surely.

Q. So that you had a definite goal in mind when you picked the one you did?

A. Surely.

Q. Now, let me ask you this: Taking any general formula, could you, as a chemist, write out an innumerable number of compounds, known and unknown, that would come within the terms of that formula?

A. You mean a formula like the one we have here?

Q. Yes.

A. Yes, surely.

Q. And would it be true it would be obvious and known how to make all those compounds?

A. No; I wouldn't say that necessarily follows.

Q. So, in other words, there is a considerable amount of speculation in writing the formulas, just as a formula, isn't there?

A. I don't follow you. I don't understand what you mean by "speculation." There are a number of formulas one could write.

The COURT. Substitute "alternatives" for "speculation" or "speculative."

Mr. SCHIMMEL. Alternatives may be specific and alternatives may be broad, your Honor. For example——

The COURT. Or choices.

Mr. SCHIMMEL. Or choices. So to that extent, to me, there is a lot of speculation involved. If you don't know where you are going, where you are headed for, you can merely write down an innumerable number of compounds, known and unknown. That is what I have in mind.

Whether anyone knows how to make them or not is also known or unknown. There may be years of research necessary in order to make many of these compounds, isn't that true, Doctor?

The WITNESS. Yes.

The COURT. I didn't mean to interrupt.

Mr. SCHIMMEL. That's all right, your Honor.

The COURT. Have you your answer?

Mr. SCHIMMEL. I think I have the answer to what I was driving at in this particular instance.

By Mr. SCHIMMEL:

Q. Now, when you say that any skilled chemist could derive the specific compound, such as that shown in Plaintiff's Exhibit 5, from the general formulation disclosed in the patent, and with the descriptive matter relating to the arbitrary symbols, you don't mean by that, do you, that that is the only compound that he could get out of that disclosure, do you?

A. Oh, no. There are other compounds he could be lead to also.

Mr. GRAHAM. May I object to that question? We are not contending that that is the only compound that a chemist would get out of it. We have specifically put in testimony of our own to show that compound, with the instructions, specifically will give you one, two, three, four—at least six. And Doctor Lewin himself said the fifth one of the six compounds is the one which specifically comes to the one we have in Exhibit 5.

The COURT. Well, I don't see any basis for your objection. You are all in agreement now. It seems to me that there are more than that one suggested.

Mr. GRAHAM. His question was that it suggested—I am trying to make a point. The instructions there lead only to this compound we have in example 5. That isn't the point. The instructions lead to five specific compounds, and one of the five is this one; and it is the one we happen to be claiming in this division of the case.

The COURT. And the question was asked, and the witness has said this is not the only one.

Mr. GRAHAM. Yes.

The COURT. The answer may remain.

By Mr. SCHIMMEL:

Q. May I ask you, Doctor, in connection with that general formula, and the instructions with respect to the substituents to be added at the various places in the formula, is it your opinion that it only denotes six specific compounds, such as Mr. Graham indicated?

A. Oh, no. More than six specific compounds. Six classes of compounds.

Q. And would you say that the specific compounds in those classes could run into the hundreds?

A.* * * This all revolves about what you mean by "specific." You mean are there hundreds you could arrive at?

Q. That is correct.

A. The answer is yes.

Mr. SCHIMMEL. That's all I have.

The COURT. Mr. Graham, do you have any redirect?

Mr. GRAHAM. Just a couple questions.

Redirect examination by Mr. GRAHAM:

Q. Doctor Lewin, in reading this formula, we have gone through the mechanics of reducing it to this

A. I am associate professor of Chemistry and Executive head of the Department of Chemistry of Georgetown University in Washington.

Q. Will you describe briefly your education and experience, mentioning publications and consulting work, for the Court?

A. I received my Doctor's degree in Chemistry from Columbia University in 1942. The next two years I held post-doctoral fellowships, one at Johns Hopkins University and the second at Mount Sinai Hospital in New York City.

Subsequently I was employed as a research chemist in two industrial positions, both in New Jersey, one at the Wallace and Tiernan Company; the second at the Schering Corporation.

In 1948, I accepted an appointment as associate professor of chemical pathology at Georgetown University Medical School.

In 1952 I was appointed as associate professor in the Department of Chemistry, at the same University. I have been a consultant for several industrial corporations and various facets of patent and organic and medicinal chemistry.

I have approximately twenty or so publications in these fields; possibly an equal number of patents in which I have been the inventor or coinventor.

Q. Are you familiar with the rules of organic chemical nomenclature?

A. I am.

Q. You have heard Doctor Martell's testimony on the nomenclature with formulas, and also Doctor Lewin. Do you subscribe to those principles?

A. I do.

Q. In other words, you feel too, that the statement of a structural formula will give a chemist a definitive organic name for that compound?

stage [indicating] and also of putting these acid groups in the D positions.

Now, the next step of the mechanics of developing significance in formula, as I understand your testimony, is to take in order, the substituents which are suggested in lines 42, 43, and 44 of the specifications, is that correct?

A. I am not sure about what you mean by "take in order."

Q. Does it matter what order they are taken in? They are listed in a certain order.

A. If you wanted to write them in order as listed, you would take them in that order.

Q. And the substituents would set up a class of compounds?

A. Yes.

Q. Would one of those classes in fact include—or rather, would one of those compounds coming out of that succession of type compounds, be the one we have in Example 5?

A. Yes.

Q. Thank you.

Mr. SCHIMMEL. I have no further questions.

(The witness left the stand.)

Mr. GRAHAM. I call as my next witness, Doctor Martin Rubin.

Thereupon Doctor MARTIN RUBIN was called to the stand by Counsel for Plaintiff, and first having been duly sworn, was examined, and testified as follows:

Direct examination by Mr. GRAHAM:

Q. What is your full name?

A. Martin Rubin.

Q. Will you state your address?

A. 8522 Milford Avenue, Silver Spring, Maryland.

Q. What is your occupation?

A. I do.

Q. I ask you now to turn to the collection of patents which is one of the exhibits, and turn to the U. S. Patent 2,524,218, carrying index tab number eight. And will you inspect the disclosure appearing in column 1, lines 25 through 46 of that patent?

A. All right.

Q. Will you describe briefly what is included there?

A. On line 30 we have a general formula which describes for us, with the modifications of the subsequent lines for the letters m, n, A, and D, a variety of compounds that may be obtained from this general formula.

Q. Would it be safe to say that that formula, plus the disclosure, is a condensation of several type formulas?

A. It would.

Q. In your opinion then, does that group of formulas include the compound we have formulated in Exhibit No. 5?

A. It does.

Q. Will you explain how you arrived at that conclusion? I believe there is a pad and pencil there.

A. If one starts with the general formula——

Q. That is Exhibit 3.

A. Exhibit 3—and accepts the first qualification in which n is specified to be two, this provides a $CH_2$ and $CH_2$ to be attached to the nitrogens. If one then further goes and takes the second qualification and accepts the second qualification, that m be either zero or positive integer, and starts with the relation that m is zero, the portion of the formula within the large parenthesis drops out and one is left then with the general formula given in the next exhibit, 4.

If one proceeds further and takes the next specification in which A is specified as being a hydrocarbon

group containing at least two carbon atoms, one now has $CH_2$ $CH_2$ from the nitrogens. The next qualification specifies that of a variety of possibilities, one may select the situation in which two hydrogen atoms, one or two hydrogen atoms, may be replaced by, for example, OR.

Further, there is a specification that R may be hydrogen, in which event the A formulation must become the type formula listed on this Exhibit 5.

Further, there is a general requirement that D be one of two possible structures or their salts, the first of these is the acetic acid derivative, $CH_2COOH$. Starting at this point, substituents for D of this group one arrives at the particular formula listed in number five.

Q. Then you would say that the generic formula, and the disclosure which we have just inspected, leads you directly and specifically to the compound in exhibit number five?

A. Yes; it does.

Q. You note that that is a duplicate of the compound we have shown specifically as exhibit 10 and the one that is Exhibit 15. Trace these through the other co-pending applications.

Now, in your opinion, does that generic formula, plus the accompanying disclosure in U. S. Patent 2,524,218, literally dictate this formula to a skilled chemist?

A. It does.

Q. Do you have any basis for your opinion, other than as an expert in this field, for saying that you feel this formula is dictated by the passage from this patent?

A. It seems clear to me that to a skilled expert this must be the upshot of using the specifications as delineated here. As a further test of this general

possibility I placed before one of my classes this general formula and the lines as given between number 30 and 47.

I put this on the blackboard with no particular explanation. And I asked this class, which is a rather broad group of students, representing countries and representing education at many levels, small universities and large universities, to take down this formula and write for me as an exercise, the specific compounds that this formula, with these specifications, would dictate to them.

They did this and submitted to me the results of their thinking. And based on their results, it has been my opinion that one must arrive at this conclusion from this specification and dictated qualifications as they are given here.

Q. Would you say these students are skilled chemists now?

A. No. These are persons who have varied backgrounds. Some are just out of college training and just beginning their graduate training in chemistry. Some are more advanced. None of them are, or have attained the doctorate degree, so I would not call them skilled chemists.

Mr. GRAHAM. At this point I would like to have the clerk mark for identification, first, the paper carrying the name of Benjamin H. Alexander.

(The document referred to was marked "Plaintiff's Exhibit 18" for identification.)

Mr. GRAHAM. The next exhibit is a paper carrying the signature of Lawrence Fishbein.

(The document was marked "#19" for Id.)

The next paper is by Teresamma Pinakatt.

(The document was marked "#20" for Id.)

The next paper is by Rachel Harris.

(The document was marked "#21" for Id.)

The next paper is by Albert LaFayette.
(The document was marked "#22" for Id.)
The next paper is by Patricia Grimes.
(The document was marked "#23" for Id.)
The next paper is by George Yip.
(The document was marked "#24" for Id.)
The next paper is by Jerome Eisner.
(The document was marked "#25" for Id.)
And the last one is by Patricia McMahan.
(The document was marked "#26" for Id.)

Mr. GRAHAM. These are the experimental results, and I will now hand them in the same order to Doctor Rubin for his comment.

The COURT. First, let me ask you if those are all of the papers?

The WITNESS. No, your Honor. These are, I would expect, just a little more than half of the group of papers turned in to me.

The COURT. What have you to say as to the ones that were not presented?

The WITNESS. They run the same pattern, your Honor. As a matter of fact, the astonishing thing is all of these papers show very much the same pattern of thinking, and I believe that is the point.

The COURT. Yes.

Mr. SCHIMMEL. I am going to object to them, your Honor. I think if we are going to have the story then we ought to have the whole story, not just a selected portion of the story. I would like to look at all of them.

Mr. GRAHAM. These are not being offered as papers of experts; something less than expert.

The COURT. I understand. Are the other papers that you have not produced available?

The WITNESS. Yes.

Mr. GRAHAM. I have them here.

The COURT. You have no objection?

Mr. GRAHAM. I will introduce them all. These are the better ones. In any class you have a spread of skill, and these represented——

The COURT. I see no reason why a few might not be selected if the witness says they are representative of all. But if opposing counsel wish to examine all, I think he should be given the opportunity.

Mr. GRAHAM. We will let him have them.

Mr. SCHIMMEL. Since this is being presented as a class exercise, we should have the class expression; not merely shall we say the one third of the class that might be A students, or one third of the class that may be B students. Let's take a look at the whole picture before we try to evaluate it.

The COURT. I have said all I wish to say at this time, and after you have cross examined, if you have some questions I will consider them.

Mr. SCHIMMEL. All right, sir.

Mr. GRAHAM. We have demonstrated, your Honor, what this passage means to somebody who is skilled in the art. The purpose of this is to show it is clear enough not only to tell somebody skilled in the art what we mean, but to others who are in training to become skilled.

The COURT. I understand. I understand the purpose and I hope it is probative.

By Mr. GRAHAM:

Q. Now, will you describe the papers, Doctor?

A. This is the paper of Mr. Benjamin Alexander. He lists the formula as given in Exhibit 3. He further says "When n is two, and m equals zero," these would be the two first requirements of this descriptive text. And comes up with formula listed as number four in this text. Then, he proceeds to go

ahead and make the possible variations that may be derived from this, using the specifications as given in the subsequent lines, 35 through 40 odd. He proceeds through this rather systematically, and comes up with several classes of formulas, several specific compounds, that is. And as number eight, he lists the compound given on the right in which he notes, in form of shorthand, I presume, again that X is equal to one, R is equal to Alkyl or hydrogen, and D is equal to acetic, which is the general formula as given here for number 5, with the stipulations.

I believe that is about the point in this particular paper.

Q. I hand you now the paper of Lawrence Fishbein for such comment as you may make on it.

A. Mr. Fishbein again starts with the formula I gave and specifies again that m equals zero and n is equal to 2, and comes up directly with the general formula listed in Exhibit No. 4. Then he proceeds to make some suggestions, and then begins to write specific compounds that may be drawn from this general formula. He does this, and on the third page, on the left hand column, second from the bottom, he comes up with the specific compound in the form of its salt. This would be the compound here [indicating].

Q. That would be the one in Exhibit 5?

A. Yes.

Q. Will you mark that one?

Mr. SCHIMMEL. Which one is that?

The WITNESS. Mr. Fishbein's paper. And the formula is on page three in the left hand column, one from the bottom. It is the specific formula of Exhibit 5.

By Mr. GRAHAM:

Q. I hand you now the paper of Teresamma Pinakatt.

A. This young lady has made no general descriptive terminology. She has gone right ahead and written specific compounds, and I note that as number 12 in her list of 19 compounds she arrives specifically at the Exhibit No. 5 in the form again of its sodium salt. This would be number 12 of her group of 19 compounds.

Q. Would you mark it?

A. Which is specifically the compound under consideration here.

Q. I hand you now the paper of Rachel Harris.

A. Miss Harris notes here, this is in response to my problem. And again she goes to specific compounds. Miss Harris has an artistic frame of mind, and I notice in formula four, she has the exact compound, except that instead of putting both the $CH_2COOH$, she put one at the top and one at the bottom and correspondingly inverted the $CH_2COOH$ in number four. But is is the exact same compound chemically.

Q. I ask you to comment on the paper of Albert LaFayette.

A. Mr. LaFayette, I notice in his very first description, decides that he is only only going to write one half of the whole formula and equilibrate A to X, so that what he writes, the right hand side of the molecule carries on the left hand side of the molecule. It is a symmetric molecule he says. Then he goes ahead further and specifically in the third try, he comes up with the exact compound which we have listed here as exhibit No. 5.

Q. Now, I hand you the paper of Patricia Grimes for comment.

A. Miss Grimes starts with the general formula given in Exhibit 3, and in her first qualification suggests that m be taken as zero and n equal two. She then says this reduces to formula in Exhibit 4. Then, she makes the following note in the next line, that for every possible variation of A, there exists the following variation in D as the acetic acid, which is the first possibility. She proceeds further then, and at the bottom of page one, she further specifies that A, the other variable, be taken as $CH_2CH_2OR$, which is next to the bottom of the parenthetical group she lists on the right as her contribution.

Q. I hand you now the paper of George Yip.

A. Mr. Yip, I note, also starts with the general formula, which he proceeds to write, in an intermediate modification of his own, but then according to the specifications proceeds, calling m zero and n equal to two, and reduces to formula in Exhibit 4. Proceeding then from this general formula, Mr. Yip arrives at one, two, three formulas, which, if we take his qualification of R equal to alkyl or hydrogen, reduces then to the specific compound in the group under consideration.

Q. I hand you a paper now of Jerome Eisner.

A. Mr. Eisner again starts with the general formula in Exhibit 3. He takes the first qualification of m equaling zero and n equal to two. And then he proceeds to write specific compounds.

He systematically goes through the variables and comes up on the second page with the one, two, three, four, five compounds down, as being the one under consideration this afternoon.

Q. That would be the one in Exhibit 5?

A. In Exhibit 5.

Q. I hand you now the paper of Miss McMahan.

A. Miss McMahan again starts with the general

formula of Exhibit 3, and taking m as zero and n equal to two, as specified, immediately comes to the next general formula in Exhibit 4. Then she proceeds to make systematic modifications in the various substituents as specified, and on the back of the first page, on the upper left, the formula in the upper left is the exact formula of Exhibit 5. It is doubled over in Miss McMahan's terminology. She says this is a symmetric molecule which is doubled over.

Q. She saved time by writing only half of it and indicating that the other half is this?

A. Yes; that is indicated here with the various of the dotted line.

Q. That is a convention chemists frequently adopt, where they have a molecule that is symmetrical they write this much of it and they say the rest of it is the other half.

The COURT. There is some variation of expression even with your symbolic language.

Mr. GRAHAM. That is part of the art of chemistry.

I would like now to interrupt Doctor Rubin's testimony to have these students individually, positively identify these papers. And then we will terminate Doctor Rubin's testimony.

The COURT. Very well, if that is necessary.

Mr. SCHIMMEL. I don't believe that I would require that, your Honor. I think I am willing to take Doctor Rubin's statement with respect to them, subject to some questions I would like to ask him as to the manner and procedure with respect to giving that examination. But I don't think it would be necessary to call each one of them individually if I can see all of the papers.

Mr. GRAHAM. It wouldn't take but a minute. The students are here. I think they have been waiting for the opportunity all afternoon.

Mr. SCHIMMEL. That is all right with me.

The COURT. I don't want to deprive them of their opportunity.

Mr. GRAHAM. Will you step down for a moment, Doctor Rubin?

(The witness left the stand.)

Will you call Mr. Benjamin H. Alexander, please?

The CLERK. Perhaps I could swear all the witnesses at once.

Mr. GRAHAM. To save time and procedure, the clerk will swear you all at one time.

Mr. SCHIMMEL. Can I ask them all if they understand the question of the oath?

The COURT. I take it that students would know that.

Mr. SCHIMMEL. I was just making the suggestion.

The COURT. I will ask first do these witnesses understand the significance of the oath they are about to take, that you are swearing before God that what you will say is the truth?

(Affirmative answers from all witnesses.)

Very well.

The CLERK. Will each of you raise your right hand.

(Thereupon, all witnesses were duly sworn.)

Mr. GRAHAM. The first witness will be Mr. Alexander.

Thereupon BENJAMIN H. ALEXANDER was called to the stand as a witness by counsel for Plaintiff, and having already been sworn, was examined, and testified as follows:

Direct examination by Mr. GRAHAM:

Q. Mr. Alexander, I hand you now a paper marked Plaintiff's Exhibit 18 for identification. Will you identify the paper?

A. This is my paper.

Q. Is it your report to Doctor Rubin based on this quiz he gave the class?

A. It is.

Q. You are one of Doctor Rubin's students?

A. I am.

Q. Did you have any particular difficulty with the question?

A. None that I recall.

Q. Thank you.

Mr. GRAHAM. I ask that this paper be marked and received in evidence.

The COURT. Received.

(The document referred to, previously marked "Plaintiff's Exhibit 18," was received in evidence.)

The COURT. Do you wish to cross-examine?

Mr. SCHIMMEL. One question, your Honor.

Cross-examination by Mr. SCHIMMEL:

Q. I would like to know from the witness whether he understood that he was to write all possible formulations under the compound, under the general formula given. Was that your understanding of the problem?

A. Would you repeat that question?

Q. Was it your understanding of the problem from the Doctor who gave you that, you were to write all possible compounds which came within the formula expression?

A. That was my understanding.

Q. That's all.

(The witness left the stand.)

Thereupon LAWRENCE FISHBEIN was called to the stand by counsel for Plaintiff, and having been previously duly sworn, was examined, and testified as follows:

Direct examination by Mr. GRAHAM:

Q. Are you one of Doctor Rubin's students?

A. Yes.

Q. I hand you now a paper marked Plaintiff's Exhibit 19 for identification, and will you identify that?

A. This is the paper that I submitted.

Q. Are you sure that is your paper?

A. Yes.

Mr. GRAHAM. I ask this be received in evidence.

The COURT. Received.

(The document marked "Plaintiff's Exhibit 19 "was received in evidence.)

Mr. SCHIMMEL. No cross examination. Oh, there is one question.

Cross-examination by Mr. SCHIMMEL:

Q. I notice on your paper that you mention 36 structures can be drawn for each of the remaining alkali metals substituted derivatives, or a total of 180 structures.

A. If one accomplishes the generic class of alkali metals there would be a number of them.

Q. Was it your understanding at the time the question was propounded, that you were to write as many specific compounds as there could possible be envisioned under that general formula?

A. Yes.

Q. Now, why did you ignore then the definition, or the designation, that n should be three?

A. Well, it wasn't ignored in a sense that the possibility couldn't exist of compounds, but in a more practical light, it didn't seem reasonable for me to.

Q. Was the question practicality, or was it just theoretical possibility?

A. Theoretically; yes.

Q. But you have not designated any compound in which n was three, have you?

A. No; I didn't; but there is a possibility.

Q. It was your understanding you would envision all possibilities?

A. Yes; but there would be an inexhaustible supply of possibilities.

Q. Any limitations put on the number that you should come up with?

A. No.

Q. Well then, would you say that you had completely answered the problem?

A. Well, to my own satisfaction I would say that I had answered the problem.

Q. Even though you didn't take into account all of the designated variables?

A. I took in the designated variables for the integer n equal two, but that satisfied my own——

Q. But you knew there were possibilities where n could be three?

A. Yes.

Q. But you didn't put those down?

A. No.

Q. How could you say that you had completely answered the problem to your own satisfaction when you knew there were possibilities with three?

A. Well, I drew the structures I thought would encompass completely for one integer, and to my own rationalization it wasn't necessary to do this for every single integer * * *

Q. Wasn't the problem to get as many as possible?

Mr. Graham. I object. It is obvious, I think I said earlier, to exhaust this formula would lead to an infinite number of compounds, and the witness who is not here as an expert, but merely as a student in training, has given the very logical answer; that he took the type, the typical integer and so as not to make his work repetitive by exploring additional ones.

The probative value is that he took it through the very steps we say that a person skilled in the art would take it.

The COURT. And it seems to me that it has been made quite clear that there are other possibilities, at least theoretically.

Mr. SCHIMMEL. I was trying to find out, your Honor, whether the witness understood the problem that was propounded to him, and if he did understand it, why there was a deliberate, shall we say, ignoring of a limitation in the problem. That was the only thing.

Mr. GRAHAM. I would suggest that type of cross examination be made of Doctor Rubin, because he is the man we are presenting as expert. And these students are merely as exhibits.

Mr. SCHIMMEL. It was the students who were to understand the question and give the answers, not Doctor Rubin. Doctor Rubin hasn't given any of his answers.

The COURT. You have no further questions?

Mr. SCHIMMEL. No.

(The witness left the stand.)

Thereupon TERESAMMA PINAKATT was called to the stand by counsel for Plaintiff, and having been previously duly sworn, was examined and testified as follows:

Direct examination by Mr. GRAHAM:

Q. Are you one of Doctor Rubin's students?

A. Yes.

Q. I hand you now a paper marked Plaintiff's Exhibit number 20 for identification, and will you identify it?

A. Yes; this is my paper. I wrote this.

Q. That paper then is in your handwriting?

A. Yes; it is.

Q. How long have you studied chemistry?

A. I received my B. S. in India and then I came here to study medicine, but they wanted me to take some more chemistry, so I enrolled in Georgetown University as special student to get in chemistry for my medical studies.

Q. In other words, they felt you were insufficiently prepared in chemistry for medical study?

A. Yes.

Q. I have no further questions.

Mr. SCHIMMEL. No questions.

(The witness left the stand.)

Mr. GRAHAM. I ask this paper, and also Mr. Fishbein's paper be received in evidence.

The COURT. Received.

(Plaintiff's Exhibits 19 and 20 were received in evidence.)

Thereupon RACHEL HARRIS was called to the stand by counsel for Plaintiff, and having been previously sworn, was examined, and testified as follows:

Direct examination by Mr. GRAHAM:

Q. Are you one of Doctor Rubin's students?

A. Yes; I am.

Q. I hand you now a paper marked Plaintiff's Exhibit 21 for identification. Is that your paper?

A. Yes; it is.

Q. Is formula number four on your paper the same as the one we have posted on the board marked Plaintiff's Exhibit 5?

A. Yes; it is.

Q. I have no further questions.

The COURT. Have you any questions of Miss Harris?

Mr. SCHIMMEL. No; I have none.

Mr. GRAHAM. I ask that Plaintiff's No. 21 be received in evidence.

The COURT. Received.

(Witness excused.)

(Plaintiff's Exhibit 21 was received in evidence.)

Thereupon ALBERT LAFAYETTE was called to the stand by counsel for Plaintiff, and having been previously sworn, was examined, and testified as follows:

Direct examination by Mr. GRAHAM:

Q. Are you one of Doctor Rubin's students?

A. Yes; I am.

Q. I hand you a paper marked Plaintiff's Exhibit for identification No. 22. Will you identify that?

A. This is my paper.

Q. I invite your attention to the formula you have numbered as three, in your list. Is that the same chemical compound as the one we have posted on the board as Number Five Exhibit?

A. Yes; it is.

Q. Thank you. I have no further questions.

Mr. SCHIMMEL. No questions.

The COURT. Where did you study prior to your study at Georgetown?

The WITNESS. I graduated from Morgan State College in Baltimore, Maryland, with a Bachelor's degree.

The COURT. Were you born in this country?

The WITNESS. Yes; I was.

(The witness left the stand.)

Mr. GRAHAM. I offer Plaintiff's Exhibit 22 in evidence.

The COURT. Received.

(Plaintiff's Exhibit 22 received in evidence.)

Mr. GRAHAM. Miss Patricia Grimes could not be here, so if there are no objections I will ask her paper be introduced in evidence.

Mr. SCHIMMEL. I have no objections.

The COURT. Received.

(Plaintiff's Exhibit 23 received in evidence.)

Thereupon GEORGE YIP was called as a witness by counsel for Plaintiff, and having been previously sworn, was examined and testified as follows:

Direct examination by Mr. GRAHAM:

Q. You are one of Doctor Rubin's students?

A. I am.

Q. I hand you a paper marked Plaintiff's Exhibit for identification No. 24. Is that your paper?

A. Yes; it is.

Q. I will invite your attention to the second formula from the bottom in your paper, first page. Is that the same compound as we have noted in our Exhibit five on the board?

A. Essentially, yes.

Mr. SCHIMMEL. What did he say?

Mr. GRAHAM. He said "essentially, yes."

By Mr. GRAHAM:

Q. I will ask him to explain that. In other words, the formula you have written is slightly generalized?

A. Yes.

Q. But, by converting the R to hydrogen, which you have noted, does it then become the compound we have?

Mr. SCHIMMEL. May I ask that all be stricken? Actually Mr. Graham is testifying.

The COURT. Yes; it is too leading, Mr. Graham.

Mr. GRAHAM. All right. I will ask the witness to explain then how the formula corresponds to the one which is Exhibit five.

The WITNESS. The only difference between this formula I have here and No. 5 is the A group I have

down here. On the side I have defined R as either alkyl group or hydrogen group.

Mr. GRAHAM. I have no further questions.

Mr. SCHIMMEL. I have one question.

Cross-examination by Mr. SCHIMMEL:

Q. You actually never did, in any of the compounds that you have on your paper, reach the formula of Exhibit 5, did you?

A. That is debatable.

Q. You mean you can't answer yes or no?

A. It can't be answered yes or no.

Q. Can you point to one which is an exact duplicate of the one on Exhibit 5 on the blackboard there?

A. On my paper?

Q. Yes.

A. As it is written down it is not the same, but if you read the definition, then you would get the same compound.

Q. That's all I have.

The COURT. Where was your education obtained prior to your entry into Georgetown?

The WITNESS. University of California.

The COURT. You were born in this country?

The WITNESS. Yes; I was, your Honor.

Thereupon JEROME EISNER was called as a witness by counsel for Plaintiff, and having already been sworn, was examined, and testified as follows:

Direct examination by Mr. GRAHAM:

Q. Are you one of Doctor Rubin's students?

A. I am.

Q. I hand you now, paper marked Plaintiff's No. 25 for identification. Will you identify this?

A. This is my handwriting and this is my paper.

Q. I invite your attention to the group of formulas in the middle of the second page. Do those formulas

correspond to what we have formulated in Exhibit five on the board?

A. Essentially they do. I have just used a shorthand——

Q. Will you explain the "essentially"?

A. There is an R on both of the top roots that can stand for alkyl group or hydrogen group. That is just a chemist's way of writing it shorter. It could have been written out in longhand.

Q. In other words, the step of substituting hydrogen for the R, is, you think, laid out in your paper?

A. It certainly is on the first page here.

Q. All right. Thank you. I have no further questions.

Cross-examination by Mr. SCHIMMEL:

Q. Isn't it true also, that the alkyl could be enlarged to quite a number of substituents also?

A. Yes, sir; that has never been debated, I don't believe. That is not the thing that is being debated from what I can gather in court.

Q. Let's not worry about that. Let's worry about the problem you were given.

A. Yes, sir; it can be a big one.

Q. So hydrogen is only one of a large number of substituents that might have been substituted for R in the formula you show here, is that correct?

A. Yes.

Q. That's all I have.

Mr. GRAHAM. I ask this paper be received in evidence as Plaintiff's Exhibit 25.

The COURT. Received.

(Plaintiff's Exhibit No. 25 was received in evidence.)

Mr. GRAHAM. Miss McMahan was not able to get here. May I ask that her paper be entered in evidence?

The Court. Received. They will all be received.

(Plaintiff's Exhibit 24 and 26 were received in evidence.)

Mr. Graham. I have a few more questions of Doctor Rubin.

Doctor Martin Rubin was recalled to the stand by counsel for Plaintiff, and testified further as follows:

Direct examination (continued) by Mr. Graham:

Q. I invite your attention to the certified record of the application before the Court. That is our Case C, Serial 215,380 and on page 10 of that record. That is the large file there.

Will you read claim two to yourself?

A. All right.

Q. Does it correspond to this formula which is exhibit fourteen?

A. Yes; it does.

Q. And does it become specifically Exhibit 15?

A. Yes; it does.

Q. Does the formula as written in the claim either in Exhibit 14 or 15 come within the disclosure of the formula we have in Exhibit 3?

A. Yes, when m is equal to zero.

Q. Thank you. No further questions.

The Court. Before you start your cross examination, we will take a short recess.

(Whereupon, a short recess was taken.)

Cross-examination by Mr. Schimmel:

Q. I don't want to embarrass you, Doctor, but in connection with the problem that you gave your class, did you try it yourself to see how many compounds you could derive out of that general formula?

A. I didn't have to try in order to determine that there would be a very large number of compounds that could be derived from this formula with the specifications as given.

Q. You know that just by looking at it?

A. That is correct.

Q. Would you probably like to hazard a guess as to how many that might be?

A. I would say in the thousands.

Q. In the thousands. So, would it be fair to say that out of a thousand possible specific compounds envisioned by that formula you might arrive at the specific compound of Plaintiff's five, which appears on the upper right hand corner of the blackboard there?

A. Yes; I would.

Q. I think I will rest on the witness there.

The Court. Do you have any further questions?

Mr. Graham. I have no further questions.

(The witness left the stand.)

Mr. Graham. We have only one further paper, and that is the stipulation which we entered into with the Patent Office concerning the specific subject matter at issue before the Court. And I now hand that to the clerk.

The Court. Received.

(The document was marked as "Plaintiff's Exhibit 27" and received in evidence.)

Mr. Schimmel. The defense has the usual exhibits, which in this particular case will be repititious of what has already been presented by the plaintiff. I was wondering whether you wanted us to go through the formality of presenting this, since everything we have here is duplicated in what has been presented.

The Court. No.

Mr. Schimmel. I don't see any necessity to burden the record with this exhibit.

The Court. I don't either.

Mr. Schimmel. So, we have nothing else to offer.

The Court. I see.

Mr. Schimmel. The evidence we have is in Plaintiff's Exhibits, the tribunals of the Patent Office, the

references relied on have all been presented in the folder, and we have agreed and stipulated that those are the references that were cited and considered in the tribunals below. So, I don't really see the necessity for encumbering the record with this.

The COURT. I don't see any necessity for it. Have you any witnesses?

Mr. SCHIMMEL. We have no witnesses to offer. We rely on the record as it was made in the Patent Office, plus the record made here by cross examination.

The COURT. I see. And will you please state for my benefit what you stated at the outset, what your objection is to the granting of the patent?

Mr. SCHIMMEL. Briefly, I think we can put the proposition this way, your Honor: Plaintiff depends on the doctrine of continuity. There is no question about that. And in order to establish his doctrine of continuity he must find in Case A, as he has indicated on his chart, a disclosure for the compound which he is seeking a monopoly on in this particular case.

He has, and we will concede, disclosed a broad general formula in that case. And from the broad general formula he says he can derive that compound.

However, the authorities say that on the basis of a generic disclosure, a broad disclosure, you cannot, and you are not entitled to a patent for a specific material which might be encompassed by that genus. The Court of Appeals in the District of Columbia, in this circuit, has specifically said so.

The COURT. You need not, unless you wish at this time, argue in detail.

Mr. SCHIMMEL. That is the issue that was raised below. If he is entitled; we say he is not entitled.

The COURT. Before you leave that point that you made, I want to ask, you maintain that he must show

the same specific thing to sustain his claim of continuity that he would have to show to obtain a patent?

Mr. SCHIMMEL. That is correct, your Honor. That is what the law is, that he has to show that in the case on which he is relying to maintain his right.

In other words, you might put it this way: He must show that he would be entitled to claim this specific compound in that first case, Case A. And the evidence shows that he did not disclose it by name. Let us go that far.

Now, the tribunals below, and the adjudications on which they rely say that has to be allowable in that case in order to entitle him to rely on that case under the doctrine of continuity. That, I think, is simply and briefly the point of this particular case.

If your Honor holds he is entitled on the basis of the disclosure in that case, then that settles the case as far as we are concerned. But we say that the adjudications and law do not permit that. And it is on that basis that the decision below refused these particular claims.

If there is any other question that your Honor has, I might clarify at this point, without arguing the case.

The COURT. No; we are not arguing now. I just wanted to clear up the issue. And I thought this was the right time to have you state it again as you did at the outset. Do you wish to make a brief reply at this time?

Mr. GRAHAM. A very brief comment. Mr. Schimmel has accurately pinpointed the issue, and that is, if the disclosure of our Case A specifically gives us the compound we have in Exhibit 5, we are certainly entitled to this patent.

Case A, as filed, contained a claim in this so-called Markush form which the Patent Office is so fond of, and one of the compounds specifically included in that

claim as a member of the group included within its scope, was exactly this compound No. 5. The formula wasn't written out in that form because the Patent Office dictates that form of disclosure.

The name doesn't appear in the specification of the patent because chemists don't normally use names of chemical compounds in their writing. In fact, chemists frequently entertain themselves with the complexity of names which can be derived for their chemical compounds.

I have here, as purely a matter of interest, a March issue of the Industrial Bulletin, published by the Arthur D. Little Company. And they have a paragraph in here in which a good example of chemical terminology appears. It would be a valuable paragraph to incorporate in a brief on argument because they identify the chemical essence, and go on to explain the full chemical name of sesame oil, and I will excuse the stenographer from trying to take this down, because it is 2–(3,4-Methylene-dioxyphenoxy)-6-(3,4-methyl-enedioxyphenyl)-cis-3,7-dioxabreyelo (3.3.0) octane.

That is the reason we don't write names of chemical compounds into our specifications.

Now, as to disclosure, the Board of Appeals in its decision of February 18, 1954, in the Patent Office, and that will be on page 80 of the certified record of the application we have before us, in the paragraph in the middle of the page there. They say right there, after reading the formula the way we asked them to that the formula will lead to the compound we have in Exhibit 5. But in the very next breath they tell us that the compound in Exhibit 5 is not disclosed in the specification.

There we have practice developed in the Patent Office which dictates using disclosure in this so-called

Markush form. The Board of Appeals admits that disclosure leads to the compound we have in Exhibit 5, and apparently they refuse a patent on the ground here that the name of the compound was not written out.

We have pointed to one of the early patents in our chain of continuity. It is number 2,428,353, in which a passage was included in this specification, specifically stating a reason why names of compounds are not used. It is simply there is enough variation in terminology, although they are definitive, variation in style is such that it is more convenient to use formulas.

The COURT. That is not the only objection, as I understand it.

Mr. SCHIMMEL. Your Honor, I have refrained from making any comment with respect to Mr. Graham's remark that our Patent Office practices and dictates this particular type of thing. There is no such thing. The statute requires the inventor to supply a full, clear, exact, and complete description of his invention.

We don't dictate anything to the inventor. All we want is a complete disclosure of what his invention is, and a particularization of what he is claiming in his invention.

The COURT. And his claim now is that anyone skilled in the art would take what is stated there and would be led to this?

Mr. SCHIMMEL. Among a thousand of compounds he would come out with that one. He might arrive at that result. He may reach it, but it doesn't expressly, it doesn't inherently reach that conclusion, because if you have one chance in a thousand, or thousands, it is difficult for me to see how you can say that it is reached, necessarily inherently and expressly by that disclosure. I just had to object to Mr. Graham

saying we dictate the type of disclosures presented. We do not. He has used the word "dictate" many times, and I have to object to it.

Mr. GRAHAM. The sole test of disclosure is what it tells a person skilled in the arts. We have presented persons skilled, experts, who are skilled in the art, and some experts in training, let us say, who came to the same conclusions uniformly time after time, and came to the same result.

If people just out of school, still in training, find the disclosure sufficient, how far do we have to go to satisfy the Patent Office?

The COURT. You admit, do you, that the degree of disclosure has to be the same to sustain your claim, to support your theory of continuity as it would be if you were claiming a patent? Do you get my question?

I wondered whether it would be argued here that you might sustain your claim of continuity even if your application at a later date was more specific in the description of the thing you are claiming a patent on. And you would not have to meet the degree of exact disclosure to sustain your claim of continuity that you would have had to show if you had asked for a patent on this specific thing.

Mr. GRAHAM. That is just what happened. Because the patent Case No. A, as filed, certified record will show, that it contained a claim in the very form of the disclosure we have been analyzing, claim one of the patent as filed.

There, the Patent Office practically required division, and that claim was carried over into another case. And that is why we reach to this one for our chain of continuity, that is, Case A. The initial formulation of the claim which leads to the compound we have in Exhibit 5 was in that patent. And that is why we point to it as having the disclosure and the

claim which justifies this current argument on the continuity.

The COURT. I don't know that you are in agreement as to what the issue is. I am trying to narrow it down. From what you say, I take it you are claiming that the disclosure in your A case, because it embodies, in a broad indefinite way, what you show in Exhibit 5 and because it was carried over, not allowed in that patent, but carried over in subsequent clarifications, has been continued and has never been dropped.

Mr. GRAHAM. We are maintaining it has been continuous and never been dropped, but I would like to modify one of your statements. It was initially embodied in Case A, not in a broad indefinite sense, but a very positive, specific sense in which that disclosure takes you directly to the compound in Exhibit 5.

The COURT. I should have added after those adjectives, "as accused."

Mr. GRAHAM. We do concede the disclosure is broad.

The COURT. I am not deciding anything now. Before I let counsel go, I would like to be sure that I know the question and the limitations of the question, and what your contentions are. I have a right to ask counsel to clear the issue. I have often said at the end of a case, in thanking counsel for making the issue so clear, and they, in return, saying they hoped the answer was clear.

I am not asking for the answer. I just want to be sure I know what your contention is. And your contention is that you can't claim continuity of a thing which was not specific enough at the outset to warrant a patent being given then.

And I understand you to say that—I want to avoid those words that you dislike, but you must concede that your A Case, now accused of being not specific

enough, and not disclosing this exact thing—I know you deny that?

Mr. GRAHAM. Yes; the fact the Patent Office record shows definitely because they never raised that question in the office. They merely require the claim of compound be divided out and carried over in another case.

The COURT. Let me ask you this question: If you were back at the beginning of A, would you say that what was disclosed there would have entitled you to a patent on what you ask now in this disputed application?

Mr. GRAHAM. Yes.

The COURT. Would you?

Mr. SCHIMMEL. We say no, your Honor. And we have got authorities to support us. I say that is the issue of this case. Could the claim that he is seeking now have been allowed in that Case A. We say no.

Mr. GRAHAM. The claim would have been allowed, and I say equally positively it could have been allowed.

The COURT. Let me ask the next question. If the Court should find that it was not stated specifically enough, described with enough exactitude to warrant a patent on that application, would you say that it was sufficiently described to be a basis for your claim of continuity?

Mr. GRAHAM. That is all the law requires. And it was described at least that well.

The law doesn't require us to maintain continuity of claimed subject matter. We have demonstrated that here because we had it. The law merely requires that if your disclosure initially was sufficient to have supported a claim to the compound you want, you are entitled to it in the continuing application.

Now, here we maintain we not only had disclosure sufficient in our case A, but we also had the claim there.

The COURT. Well, there is the difference that I anticipated.

Mr. SCHIMMEL. There is the difference, your Honor, on the question of what the law requires, and what the doctrine of continuity requires. I disagree with Mr. Graham. The foundation of the law of the doctrine of continuity recites the fact there must be the same invention. And that the basic proposition is, as we see it again, that the claim that is now out could have been allowed, and was completely disclosed in just so many terms and words in the original case on which they are claiming continuity.

The COURT. Your claim is because it wasn't——

Mr. SCHIMMEL. It was not.

The COURT. He can't have a claim of continuity. And his answer is it was sufficiently described to warrant a patent at that time, but even if it were not sufficiently described it was sufficiently embodied in the general claim and described in general to form a basis for the claim of continuity. Is that right?

Mr. GRAHAM. That's right.

The COURT. There are two angles.

Mr. SCHIMMEL. There are two angles.

The COURT. Now, how do you want to proceed with the rest? There is a lot here to be considered. Do you wish to file written briefs?

Mr. SCHIMMEL. I think we should, your Honor. Because the basic problem of law involved here, I think, requires more investigation and presentation than we have had an opportunity to present to you, or could have possibly presented by oral argument. I think it would require a written brief on those points.

The COURT. I would think so. It is very helpful to a judge, because you are dealing with terminology that is new to the Court. That is the great burden of a judge hearing a patent case. You are presenting a problem and I have to become familiar with the terminology before I can dispose of it. And you are thoroughly familiar with it. It has been your life for years. It is a burden both ways. It is my burden in the end, but I am very sympathetic with the burden that you have.

It is a little as if Einstein were asked to present his theory of relativity to the mayor of the city, or to the city council. It is a new field. But that is the law we have and that is the burden that we all have to accept. But I say that merely to assure you that I would appreciate written briefs. They need not be elaborate. I would like to have the facts and the law applicable to those facts.

How much time do you want. The issue is so clear that I think you could both file at the same time.

Mr. SCHIMMEL. I was going to suggest that, your Honor. No need for a reply brief.

The COURT. If you find there is a need, and you make application for time, I will grant it.

Mr. GRAHAM. I think it logical that we have the record before we prepare the brief.

The COURT. Very well. How soon could you give them a record, Mr. Reporter.

The REPORTER. Approximately two weeks.

Mr. GRAHAM. Let's take 20 days after receipt of the record.

The COURT. I am in no hurry. I don't wish to rush you about it.

Mr. SCHIMMEL. Thirty days after the record is ready then. Will that be agreeable?

The COURT. Yes.

Mr. Schimmel. We will both file simultaneously.

The Court. I have enjoyed the presentation of the case in spite of the difficulties that I emphasized. I commend counsel for their preparation and presentation.

Court may be adjourned until tomorrow morning at 10 o'clock.

(Whereupon, at 4:00 o'clock p. m., the above-entitled matter was adjourned.)

121 United States District Court for the District of Columbia

Civil Action No. 1637–54

Frederick C. Bersworth and the Dow Chemical Company, plaintiffs

*v.*

Robert C. Watson, Commissioner of Patents, defendant

***Memorandum by the Court***

Filed June 7, 1956. Harry M. Hull, Clerk.

Wilkin, *District Judge (by Designation)*:

This action was brought under the provisions of Section 145 of Title 35, United States Code. The plaintiffs seek a judgment authorizing the defendant, Commissioner of Patents, to grant plaintiffs letters patent upon certain claims of plaintiffs' application, Serial No. 215,380, filed March 13, 1951, entitled "Hydroxy Derivatives of Alkylene Polyamines." The complaint states that the claims are numbered 1 through 4, but since plaintiffs entered a voluntary dismissal, without prejudice, of claims 1 and 3, only claims 2 and 4 are before the court.

The specification of the application indicates that the subject matter relates to hydroxy derivatives of alkylene polyamines, a group or class of organic compounds having wide utility in the chemical arts, and particularly as sequestering agents (chelating agents) in aqueous solutions for the metal ions invariably present therein. It is not necessary to evaluate the details of the chemistry of the compounds covered by the claim, because the Patent Office has conceded that the claim defines a patentable invention.

125 The fundamental issue in the case is:

Does applicant's United States Patent 2,524,218 issued October 3, 1950 on an application filed March 28, 1946, include disclosure of subject matter of the claim before the Court sufficient to support continuity of that subject matter before the Patent Office?

The trial proceeded on the single issue of whether the applicant had sufficiently disclosed his subject matter in the parent Patent 2,524,218 to support continuity of subject matter in the application now before the Court. The Patent Office tribunals held that the application resulting in Patent No. 2,524,218 did not have a disclosure adequate to support the present claims.

The doctrine of continuity holds that a second application should be considered a continuation of a first application and entitled to the benefit of the filing date of the first application, where a patent is still sought for the substance of the invention as originally claimed. In order that the doctrine be applicable, it is necessary that the description in both applications be sufficient to support the claims. *Godrey* v. *Eames*, 68 U. S. 317; *Durgea* v. *Rice*, 28 App. D. C. 423.

It seemed to be conceded by the parties that plaintiffs' case would stand or fall on the question whether

the prior application contained a disclosure sufficient to support claims 2 and 4 of the application before the Court.

The disclosure of Patent 2,524,218 is in a concentrated form which, plaintiffs assert, sets forth specifically the formula for the elected claim of the application before the Court. Column 1, lines 25–46,
126 in the language of the chemist, and in the form required by the Patent Office, gives the detailed information defining the metes and bounds of that disclosure. The defendant conceded that if the claims at issue were directed to the general formulation, the doctrine of continuity would permit plaintiffs to rely upon the earlier patent for their date of invention, but defendant contended that the claims at issue here are not a general formulation, but instead are specific to two distinct compounds, and that such compounds were not disclosed in the earlier application.

Plaintiffs, on the other hand, contend that the claim before the Court was directly divided out of claim 1 of the earlier application. They admit that the formula presented disclosure of possibly a limitless number of other compounds, but they insist that that fact does not prevent the plaintiffs' contention that the applicant had a clear conception of the invention claimed here. They insist that the original formula embraced the specific concept presented in this case.

The statutory requirement is that the disclosure be cast "in such full, clear concise and exact terms as to enable any person skilled in the art to which it pertains * * * to make and use the same." It was conceded that the test would be what the disclosure meant to "any person skilled in the art." The plain-

tiffs, therefore, called expert witnesses and members of a class of advanced students of chemistry to support their contention that the specific directions given in the chemical formula, and the explanatory language used to define the various symbols set out in the general formula, would lead the expert to
127 the specific compounds defined in the claims at issue. The experts who testified and the members of the chemistry class, who had been given the problem as a class exercise, all stated that the specific compounds of the claims at issue were derivable from the general formula.

The evidence before this Court was clear and convincing that the symbols, when interpreted in the light of the specifications, did lead those experienced in this field of chemistry and advanced students of such chemistry to the embodiment claimed by the plaintiffs. The testimony of the experts was clear and convincing that the language of the original application, including the chemical symbols, did lead directly to the composition for which the plaintiffs seek a patent monopoly.

The defendant offered no testimony, but relied upon the material contained in the file wrapper and the opinions of the Examiner and Board of Appeals.

The Court is constrained to find in favor of the plaintiffs because the evidence sustains the plaintiffs' contention.

Findings, conclusions, and order may be prepared and submitted by counsel for plaintiff.

R. N. WILKIN,
*U. S. District Judge.*

WASHINGTON, D. C., June 6, 1956.

128 In the United States District Court for the District of Columbia

Civil Action No. 1637—54

FREDERICK C. BERSWORTH AND THE DOW CHEMICAL COMPANY, PLAINTIFFS

*vs.*

ROBERT C. WATSON, COMMISSIONER OF PATENTS, DEFENDANT

## *Findings of fact*

Filed June 29, 1956. Harry M. Hull, Clerk.

1. This is a Civil Action brought under the provisions of 35 U. S. C. 145 in which the plaintiffs, Frederick C. Bersworth, the inventor, and The Dow Chemical Co., the assignee, seek a judgment authorizing the Commissioner of Patents to issue a patent upon United States Patent Application S. N. 215,380 filed March 13, 1951 for "Hydroxy Derivatives of Alkylene Polyamines".

2. The Dow Chemical Company, a Delaware Corporation, having its offices and principal place of business in Midland, Michigan, having acquired the entire right, title, and interest in and to the patent application, was made a party coplaintiff pursuant to an Order of the Court dated February 21, 1955.

3. United States Patent Application S. N. 215,380 was filed March 13, 1951 specifically as a continuation-in-part of United States Patent Application S. N. 39,377, abandoned March 21, 1951, and was copending therewith.

129 4. United States Patent Application S. N. 39,377 was filed July 17, 1948 as a continuation-in-part of United States Patent 2,524,218 which issued October 3, 1950 on an application S. N. 657,893, filed March 28, 1946, and was copending therewith.

5. United States Patent 2,524,218, was copending with United States Patent 2,413,856 issued January 7, 1947, on application S. N. 495,207 filed July 17, 1943, and also was copending with United States Patent 2,428,353, issued October 7, 1947, on application S. N. 491,670 filed June 21, 1943, and incorporates the disclosure of said patents into its specification.

6. The Patent Office in the Board of Appeals' Decision of February 18, 1954, refused to allow the claims of application S. N. 215,380 as being anticipated by United States Patent 2,428,353 and 2,413,856 on the ground that the disclosure of United States Patent 2,524,218 was insufficient to maintain continuity of the subject matter before the Patent Office to give the application S. N. 215,380 the benefit of the filing date of United States Patent 2,407,645.

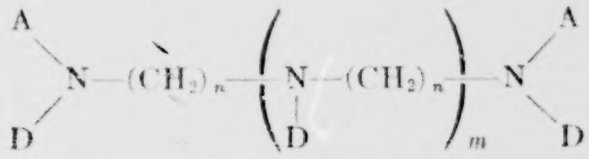
7. United States Patents 2,407,645 and 2,428,353, having been filed simultaneously by the same inventor and having been cross referenced effectively make one large disclosure.

8. United States Patent 2,413,856 issued January 7, 1947 on application S. N. 495,207 filed July 17, 1943, was copending with United States Patents 2,407,645 and 2,428,353.

130 9. The disclosure of United States Patent 2,524,218 included a clear description of the concept of the subject matter of the application before the Court, and refers to United States Patent 2,407,645, which in turn refers to the application S. N. 491,670 which became United States Patent 2,428,353.

10. A portion of the disclosure of United States Patent 2,524,218 is in a concentrated form, commonly used in specifications and accepted by the Patent Office, and includes the following passage at column 1, lines 25–46:

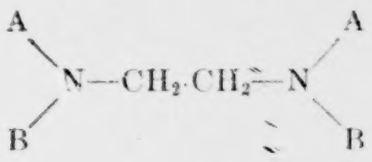
The alkylene polyamine derivatives of the present invention are represented by the following formula:



wherein n is either 2 or 3 and m is either zero or a positive integer and wherein D is a member of the group consisting of—$CH_2COOH$, —$CH_2CH_2COOH$, and their alkali metal salts, and wherein A is a hydrocarbon group containing at least two carbon atoms and having not more than two hydrogen atoms replaced by a member of the group consisting of—$(CH_2)_2COOR$,—$(CH_2)_2COOM$,—$SO_3H$,—$SO_3M$,—OR and OR′OR wherein R is alkyl or hydrogen, M is an alkali metal, and R′ is alkylene, said groups being spaced at least two carbon atoms from the nitrogen atoms.

11. Claim 2 of United States Patent application S. N. 215,380 was the species elected for prose-
131 cution before the Patent Office and it reads as follows:

2. The compound conforming to the formula



wherein A is a substituent group selected from the group consisting of ethanol and propanol and B is an acetic acid substituent group.

Claim 4 defines the basic metal salts of the compounds of Claim 2.

12. The evidence before this Court was clear and convincing that the symbols set out in the formula as in finding 10, when interpreted in the light of the

specifications, did lead those experienced in this field of chemistry and advanced students of such chemistry to the embodiment claimed by the plaintiffs. The testimony of the experts was clear and convincing that the language of the original application, including the chemical symbols, did lead directly to the composition for which the plantiffs seek a patent monopoly.

13. Claim 2 of United States Patent Application S. N. 215,380 is specifically supported by the disclosure of United States Patent 2,524,218. In fact, Claim 2 of S. N. 215,380 was directly derived from Claim 1 of United States Patent 2,524,218 as filed.

132 14. The Patent Office having conceded that the subject matter of United States Patent Application S. N. 215,380 is inventive, claims 2 and 4 define inventive subject matter.

15. Claims 1 and 3, directed to nonelected subject matter, were withdrawn from consideration by stipulation of the parties.

*Conclusions of Law*

1. The application S. N. 215,380 was a proper continuation-in-part of S. N. 39,377.

2. Application S. N. 39,377 was a proper continuation-in-part of United States Patent 2,524,218.

3. Application S. N. 215,380 is accordingly entitled to the benefit of the filing date of United States Patent 2,524,218.

4. Certain generic disclosure of United States Patent 2,524,218 appearing at column 1, lines 25–46, being in the Markush form, which is a creature of Patent Office practice, is a part of the total disclosure of the Patent and should be considered specific support for those compounds to which the disclosure specifically leads.

5. Claims 2 and 4 are not anticipated by the prior art cited, they are patentable, and patent may issue on the application S. N. 215,380.

6. The doctrine of continuity of subject matter before the Patent Office requires that United
133 States Patent application S. N. 215,380, which defines a concept initially disclosed in United States Patents 2,407,645 and 2,428,353, and which was carried in substance consistently through United States Patent 2,524,218 and United States Patent application S. N. 39,377, be accorded the benefit of the early filing date namely, June 21, 1943. Godfrey v. Eames, 68 U. S. 317.

R. N. Wilkin,
ROBERT N. WILKIN,
*U. S. District Judge.*

JUNE 29, 1956.

134 In the United States District Court for the District of Columbia

Civil Action No. 1637–'54

FREDERICK C. BERSWORTH AND THE DOW CHEMICAL COMPANY, PLAINTIFFS

*v.*

ROBERT C. WATSON, COMMISSIONER OF PATENTS, DEFENDANT

*Order*

Filed June 29, 1956. Harry M. Hull, Clerk.

This action came on to be heard at the last term of Court and thereupon, upon consideration thereof; it is this 29th day of June, 1956,

Ordered, adjudged and decreed that:

The Commissioner of Patents is authorized to issue a patent on application S. N. 215,380 containing claims 2 and 4.

The Complaint, insofar as it relates to claims 1 and 3 representing non-elected subject matter, is dismissed without prejudice.

The expenses incurred herein shall be borne by the plaintiffs.

R. N. Wilkin,
ROBERT N. WILKIN,
*U. S. District Judge.*

135 United States District Court for the District of Columbia

Civil Action No. 1637–54

FREDERICK C. BERSWORTH AND THE DOW CHEMICAL COMPANY, PLAINTIFFS

*v.*

ROBERT C. WATSON, COMMISSIONER OF PATENTS, DEFENDANT

*Notice of Appeal*

Filed August 28, 1956. Harry M. Hull, Clerk.

Notice is hereby given this 27th day of August, 1956, that defendant Robert C. Watson, Commissioner of Patents, appeals to the United States Court of Appeals for the District of Columbia Circuit from the Order of this Court entered on June 29, 1956, in favor of the plaintiffs, Frederick C. Bersworth and The Dow Chemical Company, against said defendant, Commissioner of Patents, said Order authorizing the defendant to issue letters patent of the United States based upon the patent application of said Frederick C.

Bersworth, Serial No. 215,380, filed March 13, 1951, and containing claims 2 and 4 thereof.

Respectfully submitted.

C. W. MOORE,
*Solicitor, U. S. Patent Office,*
*Attorney for Defendant.*

AUGUST 27, 1956.

J. SCHIMMEL,
*Of Counsel.*

United States District Court for the District of Columbia

Civil Action No. 1637–54

FREDERICK C. BERSWORTH AND THE DOW CHEMICAL COMPANY, PLAINTIFFS

*v.*

ROBERT C. WATSON, COMMISSIONER OF PATENTS, DEFENDANT

*Order To Transmit Original Exhibits*

Filed September 19, 1956. Harry M. Hull, clerk.

It is hereby ordered that the Clerk of the Court be and is hereby authorized and directed to transmit the original exhibits filed herein and designated in the Stipulated Designation of Record on Appeal as Plaintiffs' Exhibits 2 to 10, inclusive, and 12 to 26, inclusive, as part of the record on appeal, in lieu of copies thereof.

F. DICKINSON LETTS,
*Judge.*

Date: September 19, 1956.

Patented Jan. 7, 1947 2,413,856

# UNITED STATES PATENT OFFICE

2,413,856

## VINYL POLYMER PLASTICIZED WITH ETHYLENE DIAMINE TETRAACETIC ACID ESTER

Frederick C. Bersworth, Verona, N. J.

No Drawing. Application July 17, 1943, Serial No. 495,207

6 Claims. (Cl. 260—36)

1

This invention relates to the plasticizing and modifying of certain vinyl compounds of a resinous nature, and more particularly to the incorporation of esters of polyamino-polyacetic acids as modifying agents in vinyl chloride polymers, vinyl chloride-vinyl acetate copolymers, vinyl chloride-vinylidine chloride copolymers, and vinyl butyral polymers.

In the preparation of useful products of these vinyl polymers and copolymers, it is advisable to add a softening or plasticizing agent. But the addition of such agents generally decreases the tensile strength of the finished product to a considerable degree; furthermore, each such agent decreases the tensile strength to a different degree. The plasticizing or softening agent must in any event be compatible with the polymer, and must not bleed out when the polymer is worked.

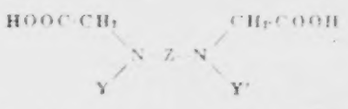
It is an object of this invention to set forth a class of plasticizers for the above-mentioned vinyl polymers and copolymers which have much less adverse effect than do ordinary plasticizers on the tensile strengths of products made of such polymers and copolymers.

Other objects will appear in the following detailed explanation of the invention.

Briefly, these objects may be accomplished by the incorporation of an ester of a polyamino polyacetic acid with the said vinyl polymers and copolymers.

The esters which may be advantageously incorporated with the said vinyl polymers and copolymers to obtain the desired results may be defined in terms of the amino acid and in terms of the alcohol used for esterification. The method for preparing these esters is set forth in detail in my copending application, Ser. No. 491,670, filed June 21, 1943.

The polyamino polyacetic acid may be broadly defined by the formula:



wherein Y and Y' are the same or different and are either hydrogen, —$CH_2$·COOH, an alkyl group, a cycloalkyl group, or an alkylol group; and Z is an alkylene group, an alkylene-ether group, an alkylol group, or a polyalkylene-amine group in which latter case the amino nitrogen atoms intermediate the polyalkylene chain are attached either to a hydrogen atom or to a —$CH_2$·COOH group. Of particular value in the present invention are the esters of the amino tetra (or higher) acetic acids, i. e. of those acids which correspond to the above formula wherein Y and Y' are —$CH_2$·COOH, and also wherein Z is an alkylene or a polyalkylene-amine group (i. e. >N—Z—N< is an alkylene-polyamine radical or a polyalkylene-polyamine radical).



2

Esters formed from any alcohol containing at least one esterifiable —OH group attached to a carbon atom which is not part of a benzene ring may be used advantageously. The esterification, however, is preferably complete as to all the free carboxylic acid groups of the amino acid, and must be complete as to at least two of them and there must, by definition, be at least two such free carboxylic acid groups. The esters used in the present process will therefore correspond to the foregoing formula except for the replacement of hydrogens of the carboxylic acid groups —COOH by R—, R— being the radical of an alcohol ROH.

The resinous vinyl compounds with which the above defined esters have a beneficial plasticizing effect include the polyvinyl chloride polymers and the polyvinyl chloride copolymers. Specifically included are the straight polymers

$$—(—CH_2—CHCl—)_n—$$

such as "Koroseal," the vinyl acetate-vinyl chloride copolymers, also called polyvinyl chloride-acetate, the vinyl chloride-vinylidine chloride copolymers, and polyvinyl butyral.

The following is a general method for incorporating the ester of the polyamino polycarboxylic acid in the resin. The vinyl resin is first dissolved in a volatile solvent thereof inert to both the resin and the ester and which is preferably capable of dissolving both substances. About a 5% solution of the resin has been found suitable for most practical operations. To this is added the ester in the desired amount which is preferably about one third of the weight of the resin.

After thorough incorporation, the resulting composition may be sheeted out and dried by evaporation of the solvent, or otherwise manipulated to form the desired final product. This composition, before evaporation of the solvent, may be fairly liquid or may be jelly-like, depending principally upon the amount of solvent used.

It is to be understood that any quantity of the ester will have a softening effect and as the quantity of ester added to a given weight of resin is increased, the softer the resin will become until it finally dissolves. The quantity of ester used will therefore be in accordance with the characteristics of the final product desired for a particular purpose, but as stated above, about ⅓ by weight of the ester based on the weight of resin will give satisfactory results for most ordinary purposes.

For evaluation and comparison purposes, the more or less jelly-like mixture of resin and ester in solution, prepared as above described, is drawn to a predetermined thickness on a glass plate, thus forming a film which will dry down to about 1/10 the thickness originally measured. In the examples which follow the films were made .03 inch

thick when in solution and were dried down to .001 inch when dry. These dry films were removed and part of them were milled on a two roll mill to ascertain the properties of the treated resin. The remainder were then tested for per cent elongation and tensile strength at these elongations after having been conditioned in a room at 65% relative humidity at 70° F. for four hours.

This invention is not limited to the preferred method of incorporating the ester. For instance, the resin and the ester may be added together and the mixture sheeted on a two-roll mill without the use of any solvent. Similarly any method which serves to incorporate the ester in the resin in a satisfactory manner is included in this invention.

In order to compare the effect of adding the esters of polyamino-polycarboxylic acids referred to herein with that of well-known plasticizers, four of the most widely used plasticizers were selected and incorporated in exactly the same manner and relative amounts as the said esters. To facilitate comparisons between the physical properties of the final products, tables are given in Examples I, II, and III in which each of the plasticizers is designated by a letter as follows:

A represents the tetrabutyl ester of ethylene diamine tetraacetic acid.
B represents dibutyl sebacate.
C represents tricresyl phosphate.
D represents triethylene glycol di-2-ethyl butyrate.
E represents triethylene glycol di-2-ethyl hexate.

### *Example I*

To 570 grams of mesityl oxide were added 30 grams of polyvinyl chloride. To the resulting solution 10 grams of the tetrabutyl ester of ethylene diamine tetraacetic acid were added. The same general procedure of forming films as outlined above was then followed. The following is a table of the results obtained on testing representative samples:

| | | A | B | C | D | E |
|---|---|---|---|---|---|---|
| Percent plasticizer [1] | | 33 | 33 | 33 | 33 | 33 |
| Tests | | | | | | |
| Tensile | lb./sq. in. | 4,800 | 2,300 | 2,000 | 1,700 | 3,300 |
| Elongation | percent | 180 | 170 | 180 | 185 | 280 |
| Compatibility | do | 100 | 100 | 100 | 100 | 100 |
| Spew | | 0 | 0 | 0 | 0 | 0 |

[1] Based on weight of resin.

### *Example II*

Following the same procedure as in Example I, polyvinyl chloride-acetate (vinyl chloride-vinyl acetate copolymer) was used as the resin. Results of tests on representative products thereof were as follows:

| | | A | B | C | D | E |
|---|---|---|---|---|---|---|
| Percent plasticizer [1] | | 33 | 33 | 33 | 33 | 33 |
| Tests | | | | | | |
| Tensile | lb./sq. in. | 3,000 | 370 | 2,400 | 600 | 1,400 |
| Elongation | percent | 320 | 320 | 280 | 335 | 365 |
| Compatibility | do | 100 | 100 | 100 | 100 | 100 |
| Spew | | 0 | 0 | 0 | 0 | 0 |

[1] Based on weight of resin.



### *Example III*

Following the same procedure as in Examples I and II, but using polyvinyl butyral as the resin, the following results were obtained in testing representative samples:

| | | A | B | C | D | E |
|---|---|---|---|---|---|---|
| Percent plasticizer [1] | | 33 | 33 | 33 | 33 | 33 |
| Tests | | | | | | |
| Tensile | lb./sq. in. | 4,800 | 3,700 | 3,600 | 3,700 | 4,700 |
| Elongation | percent | 480 | 345 | 345 | 320 | 345 |
| Compatibility | do | 100 | 100 | 100 | 100 | 100 |
| Spew | | 0 | 0 | 0 | 0 | 0 |

[1] Based on weight of resin.

### *Example IV*

Using polyvinyl chloride as the resin and the tetra-isopropyl ester of ethylene diamine tetraacetic acid as the ester, and following the same procedure of preparing and testing films as already described, the following results were obtained on testing representative samples:

| | | |
|---|---|---|
| Per cent of plasticizer [1] | | 33 |
| Tests: | | |
| Tensile | lb./sq. in. | 4,760 |
| Elongation | per cent | 200 |
| Compatibility | do | 100 |
| Spew | | None |

[1] Based on weight of resin.

From the foregoing examples, it may readily be seen that the incorporation of the herein defined esters of polyamino polycarboxylic acids with the vinyl polymers herein set forth serves not only to plasticize those polymers but also to yield a product with a tensile strength greater than that encountered when ordinary plasticizers are used.

I claim:

1. A plasticized resinous product comprising a vinyl compound selected from the group consisting of vinyl chloride homopolymers, vinyl chloride-vinyl acetate copolymers, vinyl chloride-vinylidene chloride copolymers, and vinyl butyral polymers, and a tetra-ester of ethylene diamine tetraacetic acid.

2. The product of claim 1 wherein said ester is the tetrabutyl ester of ethylene diamine tetraacetic acid.

3. A plasticized resinous product comprising a vinyl compound selected from the group consisting of vinyl chloride homopolymers, vinyl chloride-vinyl acetate copolymers, vinyl chloride-vinylidene chloride copolymers, and vinyl butyral polymers, and an ester of ethylene diamine tetraacetic acid; said ester being at least a di-ester.

4. The product according to claim 3 wherein said vinyl compound is a vinyl chloride polymer —(—$CH_2$—CH Cl—)$_x$.

5. The product according to claim 3 wherein said vinyl compound is a vinyl chloride-vinyl acetate copolymer.

6. The product according to claim 3 wherein said vinyl compound is a vinyl butyral polymer.

FREDERICK C. BERSWORTH.

Patented Oct. 7, 1947 2,428,353

# UNITED STATES PATENT OFFICE

2,428,353

## LOWER ALIPHATIC ESTERS OF ETHYLENE- AND PROPYLENE-DIAMINE N,N'-TETRA-ACETIC ACIDS

Frederick C. Bersworth, Verona, N. J.

No Drawing. Application June 21, 1943, Serial No. 491,670

8 Claims. (Cl. 260—482)

1

This invention relates to the production of amino acid esters. More particularly, it relates to a process for producing esters of polycarboxylic amino acids, and to the products so prepared.

The esterification of amino acids has always been difficult. Although some of the esters of primary alpha amino acids, such as the amino acids of the proteins, have been prepared, esters of the more complex amino acids, especially of the polycarboxylic amino acids, have not, so far as I am aware. In methods hitherto employed for esterifying amino acids, it has been customary to pass HCl gas into a mixture of the desired alcohol and amino acid as the mixture is heated. However, gaseous HCl is inconveneint to use, and the yields obtained with it are often poor. When attempts are made to esterify amino acids, particularly polycarboxylic amino acids, with a sulfuric acid catalyst in the manner used in esterifying ordinary organic acids, trouble is encountered due to the basic nature of the amino group in the amino acid. If small amounts of sulfuric acid are used, the amino groups react with or neutralize it and no esterification takes place; if large amounts of sulfuric acid are used many side reactions are likely to occur between the acid and the organic substances present, resulting in poor yield and off-color products. When the resulting esters are such that they are non-volatile and so high boiling that they cannot be distilled, the non-volatile foreign bodies introduced by these side reactions may be removed only with the greatest difficulty.

It is an object of this invention to prepare the esters of amino acids, and in particular of polycarboxylic amino acids.

Another object is to prepare these esters by a simple and easily performed process whereby good yields are obtained, with relatively little by-product formation.

Still another object is to produce esters of polycarboxylic amino acids, which esters have not been previously known and which are of particular commercial value.

Briefly, the process of this invention may be accomplished by reacting a polycarboxylic amino acid with an alcohol in the presence of sufficient strong mineral acid to form the amino acid-mineral acid addition product, the reaction being carried out so that water resulting from the esterification reaction is driven off, and at the completion of the reaction, any excess alcohol is removed by distillation, leaving the ester usually in the form of an addition product with the mineral acid. This product may then be washed with an alkaline solution to neutralize, and permit separation of the mineral acid. Alternatively, the washing step may be carried out before distilling off the alcohol.

In order to prevent undesired side reactions it

2

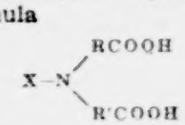
has been found preferable, although not absolutely necessary, to combine the mineral acid and the amino acid to give an addition product and then reacting this addition product with the alcohol.

The amino acids which are esterified in accordance with the procedure of the present invention are, as already stated, polycarboxylic amino acids. They may be prepared, for example, in accordance with my copending application, Serial No. 491,669, filed June 21, 1943. The particular polycarboxylic amino acids with which the procedure of the present invention is concerned are those having at least one secondary or tertiary amino nitrogen atom, and also at least two carboxyl groups —COOH, each carboxyl group being separated from every other carboxyl group in the amino acid by at least one of said secondary or tertiary amino nitrogen atoms. The prefix "poly" is used in its broad sense meaning "more than one."

The present invention is accordingly not concerned with the well-known amino acids such as glycine, leucine, cysteine, aspartic acid, histidine, and lycine, which commonly occur as structural units in the proteins and peptides. These amino acids do not fulfill the requirements set forth above.

The polycarboxylic amino acids to which the present invention refers may be represented by the general formula

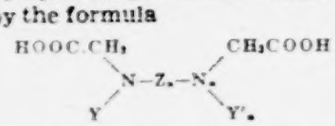
(1)



wherein R and R' are the same or different organic radicals and X is an organic radical or hydrogen. Hence X may or may not contain a carboxyl group or groups.

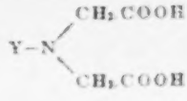
The preferred type of polycarboxylic amino acids within the above-defined group is characterized by containing at least two —$CH_2COOH$ radicals, each attached to the same or a different amino nitrogen atom in the molecule. These preferred polycarboxylic amino acids may be represented by the formula

(2)



wherein Y and Y' are the same or different and are either hydrogen, —$CH_2COOH$, an alkyl group, a cycloalkyl group, or an alkylol group; $n$ is a number not greater than 1, each $n$ being the same, and when $n$=zero, the formula becomes that of a polycarboxylic monoamino acid:

(3)



The symbol $Z_n$ in Formula 2 is either an alkylene group, an alkylene-ether group, an alkylol group, or the radical of a polyalkylene polyamine in which latter case the amino nitrogen atoms intermediate the polyalkyene polyamine chain are attached either to a hydrogen atom or to a $-CH_2COOH$ group.

Of particular interest are the esters of the amino tetra- (or higher) acetic acids—i. e., of those acids corresponding to Formula 2 wherein Y and Y' are $-CH_2COOH$ These esters, which so far as I am aware have never been prepared before, are useful notably in plastics formulation, and for other purposes.

Specific examples of the polycarboxylic amino acids which may be esterified by the process of the present invention include ethylene diamine N,N'-tetraacetic acid; propylene 1,2-diamine N,N'-tetraacetic acid; 1,3-diamino propanol-2 N,N'-tetraacetic acid; diethylene triamine N,N'-tetraacetic acid; diethylene triamine N,N',N''-pentaacetic acid; hexamethylene diamine N,N'-tetraacetic acid; cyclohexyl amine N-diacetic acid; α-butyl amine N-diacetic acid; ethylene diamine N,N'-diacetic acid; dipropylene triamine N,N'-tetraacetic acid; ethylene diamine N,N'-triacetic acid N'-ethanol;

$(HOOC-CH_2)_2NCH_2CH_2-O-CH_2-$
$O-CH_2CH_2N(CH_2COOH)_2$

Although the correct amount of any strong mineral acid which is non-injurious to the reactants under the conditions imposed is suitable in the procedure of this invention, sulfuric acid is preferred, except in cases where the alcohol is sensitive to it, as stated in more detail below. Hydrochloric acid can be used in place of sulfuric, if desired, although the reaction generally goes less effectively when hydrochloric acid is used. To be effective the acid should be stronger than phosphoric, $H_3PO_4$.

It has been found that if the pH of the amino acid-mineral acid addition product is brought to or below pH=0—that is, the strength of the mineral acid is brought to at least 1N, and preferably about 4N—the amino acid-mineral acid addition product may be crystallized from solution. The crystals formed will then always contain the correct amount of acid for the esterification process regardless of the excess acid which has been used, since any excess acid will be removed by filtration from the crystals or by equivalent treatment. In determining the amount of mineral acid which fulfills the foregoing requirement, the properties of the particular amino acid and the particular mineral acid must be considered. In general, the amount of the mineral acid must exceed the equivalent amount of amino nitrogen present in the amino acid. At least an equivalent amount is necessary to overcome the basicity of the amino nitrogen, and somewhat of an excess is necessary to prevent hydrolysis and to serve as a catalyst for the esterification reaction. In the case of ethylene diamine N,N'-tetraacetic acid and sulfuric acid, two amino nitrogen atoms are present which require two hydrogen atoms and, as sulfuric acid is bivalent, one mol of ethylene diamine N,N'-tetraacetic acid requires one mol of sulfuric acid. In actual practice considerable excess of sulfuric acid is preferably added to the amino acid to prevent hydrolysis, and the resulting crystals of amino acid-mineral acid addition product contain about 50% more acid than theoretical, i. e., one mol of ethylene diamine N,N'-tetraacetic acid forms



crystals with about 1½ mols of sulfuric acid. In addition, in an aqueous solution sufficient sulfuric acid should preferably be present so that the resulting composition is about 4 N with respect to sulfuric acid in order to assure proper crystallization and to prevent hydrolysis, though any amount in excess of this is not harmful and the mineral acid contents of the crystals are the same. Once the amino acid-mineral acid addition product has been formed, crystallized, and dried, the amount of acid in the crystals may, if desired, be determined. Between about 1.1 and 2 mols of sulfuric acid to 1 mol of ethylene diamine N,N'-tetraacetic acid will always be found present. This is the desirable range of sulfuric acid which should be present when the amino acid is reacted with the alcohol. Likewise, if a mono amino polycarboxylic acid is used, half as many mols of sulfuric acid will be needed to form the addition product. When hydrochloric acid is used instead of sulfuric, the molar amount must be doubled. In other words, the amount of mineral acid used in forming an addition product with a polycarboxylic acid is such that the number of equivalents of said mineral acid so used per mol of amino acid equal at least about 1.1 times the number of amino nitrogen atoms per molecule of said polycarboxylic amino acid.

In the preferred method the alcohol is reacted with the crystalline amino acid-mineral acid addition product formed as described above. Alternatively, the ester may be prepared by thoroughly mixing the predetermined proper amount of diluted sulfuric acid with the amino acid, and reacting the resulting composition with the alcohol, with simultaneous removal of the water of solution and the water of formation of the ester. Use of the crystals is, however, preferred for several reasons. They provide the exact composition for the reaction with the alcohol, thus eliminating possible errors; furthermore, impurities that may be in the amino acid are largely eliminated by this crystallization procedure.

Another method of preparing the crystalline amino acid-mineral acid addition product is to treat the dry crystalline amino acid with substantially anhydrous mineral acid, in the predetermined amount, for example, one mol of ethylene diamine N,N'-tetraacetic acid with 1.1 to 2 mols of sulfuric acid in the form of 96% acid. When these are mixed together, a substantially dry crystalline powder is formed. This procedure avoids the necessity for drying, and the presence of extra water.

In carrying out the process of this invention, any alcohol containing at least one esterifiable —OH group attached to a carbon atom which is not part of a benzene ring, may be used. Compounds wherein the —OH group is attached to a carbon atom which is part of a benzene ring are properly phenols rather than alcohols, and are not included within the present invention.

Certain alcohols are more or less sensitive to sulfuric acid such as some of the polyhydric alcohols, e. g., glycerine and the mono ethyl ether of ethylene glycol, and when employing them it is preferable to use a mineral acid such as hydrochloric acid, toward which these alcohols are not sensitive.

In carrying out the esterification procedure, the alcohol is mixed with the polycarboxylic amino acid and the mineral acid which are preferably in the form of the addition product as

already described, and the mixture is heated, preferably to boiling. Water is formed by the esterification reaction, and this water, together with any water initially present, passes off together with some of the alcohol (the amount of which depends upon its boiling point). The water should be removed continuously in order to prevent hydrolysis of the ester. Loss of alcohol should, however, be prevented as far as possible. Therefore the water and alcohol should be separated, the water withdrawn and the alcohol returned to the reaction mixture. When the alcohol is immiscible with water they may be separated, after condensation, by a trap. When they are miscible, any of the known methods of separating alcohol and water may be used for this purpose—for example, use of a fractionating column, chemical dehydration or salting out, or use of a water immiscible third substance.

After this esterification reaction, the mineral acid (which is usually then present as some form of addition product with the ester) is removed from association with the ester by treating the reaction with a mild alkali, e. g., sodium carbonate. The resulting salt of the alkali and the mineral acid is then separated from the ester by known methods, e. g., by washing with water. Excess alcohol (which is ordinarily present because it is preferable to employ an excess of the alcohol in making the ester when all carboxylic acid groups are to be esterified) may be removed by distillation or other suitable means; this may be done before or after the neutralization step.

In order to more fully understand the invention, but not to limit it, the following examples are given:

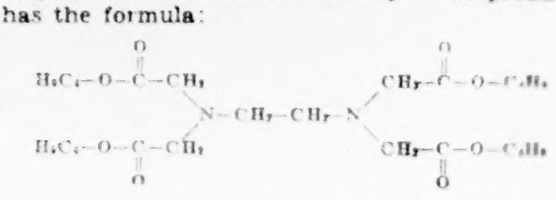
*Example I.—Preparation of the tetrabutyl ester of ethylene diamine tetraacetic acid*

*Step 1.*—Preparation of the amino acid-mineral acid addition product.

2 mols of 96% sulfuric acid diluted with 1000 cc. of water are used to dissolve ¼ mol of anhydrous ethylene diamine N,N'-tetraacetic acid at the boiling temperature of the aqueous acid. Upon cooling, the amino acid-sulfuric acid addition product is crystallized out, filtered, and dried at 110° C. Combined and free sulfuric acid is found to be about 1½ mols per mol of the amino acid, or ⅜ of a mol of sulfuric acid on the amount of amino acid used.

*Step 2.*—Esterification.

214 grams of the crystalline addition product prepared by Step 1 are mixed with 428 cc. of normal butanol and heated to boiling. Water is removed, for example by a trap reflux, as the reaction continues and when water is no longer evolved, the reaction is complete. The resulting reaction mixture is then shaken with dilute sodium carbonate solution, the aqueous layer is removed, and the excess butanol distilled off. Alternatively, the excess butanol may be distilled off before treating with the sodium carbonate. The residue is a clear, brownish-yellow liquid consisting of almost pure tetra-n-butyl ester of ethylene diamine N,N'-tetraacetic acid. Yield of product is over 90% of theory. The product has the formula:



If desired, lesser amounts of the alcohol may be used, resulting in the mono-, di-, or tri-ester or mixtures of these.

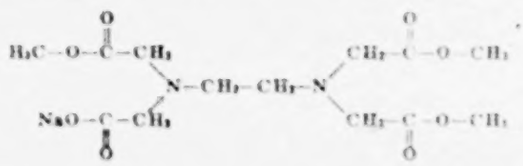
By proceeding in the manner of the foregoing example, other alcohols may be reacted with the herein-described amino polycarboxylic acids, in conjunction with strong mineral acids, to produce the corresponding esters of those amino acids.

To illustrate in further detail the foregoing statement, the following examples are given:

*Example II.—Preparation of methyl esters of ethylene diamine tetraacetic acid*

302 cc. of anhydrous methyl alcohol is mixed cold with 30 cc. of 93% $H_2SO_4$. To this is added 42 grams of ethylene diamine tetraacetic acid, and the mixture heated on a steam bath under a reflux condenser for 2 to 2½ hours. (No attempt is made to remove water introduced into the reaction with the $H_2SO_4$ or water of reaction.) The mixture is allowed to cool, and the excess alcohol filtered off. The yield is 260 grams of the ester sulfate as compared with the theoretical yield of 264 grams. The $H_2SO_4$ is then neutralized with $Na_2CO_3$ or $BaCO_3$ (this step may be performed either before or after the removal of the alcohol).

The product formed, when $Na_2CO_3$ is used to neutralize, is the monosodium salt of the trimethyl ester of ethylene diamine tetraacetic acid.



If barium carbonate is used to neutralize, the corresponding barium salt will be formed, containing of course two of the above molecules joined together through one barium atom in place of sodium.

The ester sulfate is a solid, while the ester shown in the foregoing formula is a clear mobile liquid with a boiling point so high that it cannot be distilled.

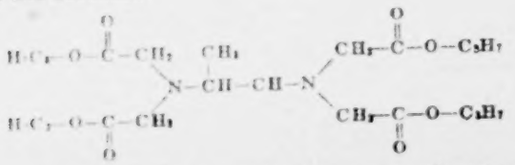
The monosodium (or other metal) salt of the trimethyl ester may be converted into the trimethyl ester of ethylene diamine tetraacetic acid by treating that salt with a mild acid, thereby converting the NaO-group to an HO-group.

It will be noted that removal of water was not necessary for the formation of the trimethyl ester, although such removal may be carried out if desired. However, to make the tetramethyl ester, water removal is necessary in the manner already described herein. Specifically, benzene or toluene may be introduced and the reaction mixture heated to remove the water.

*Example III.—Preparation of the tetraisopropyl ester of 1,2-propylene diamine tetraacetic acid*

10 grams of 1,2-propylene diamine tetraacetic acid, 6 cc. of $H_2SO_4$ 93%, and 80 cc. of anhydrous isopropyl alcohol are made into a paste. This paste, which melts at about 50° C., is heated at 60° C. for about 3 hours under a reflux condenser. The excess alcohol is removed and the acid neutralized with $Na_2CO_3$. A yield of 75%, based on the amino acid used, of the tetraisopropyl ester of 1,2-propylene diamine tetraacetic acid is obtained. The product is a viscous light colored

liquid which is somewhat soluble in water, and has the formula



The esters of this invention will accordingly be represented by the same formulas already given herein for the corresponding acids, except of course that some or all of the carboxyl groups will be replaced by ester groups —COOR'' wherein R'' is the radical of an alcohol R''OH. As shown in Example II, when not all of the carboxyl groups are replaced by ester groups, the hydrogen of the unreplaced carboxyl groups may be substituted by a metal.

The esters formed by this process are useful for several purposes. For instance, the tetra-n-butyl ester of ethylene diamine N,N'-tetraacetic acid, when compounded with synthetic or natural rubber in making tire stocks, greatly reduces the aging effect caused by the heating of tires when they are run. The tetraisopropyl ester and the tetra-n-butyl ester of ethylene diamine N,N'-tetraacetic acid are also useful plasticizing agents for polyvinyl chloride and other vinyl polymers. Furthermore, the esters described herein are generally useful plasticizers for "butyl" rubber (butadiene-isobutylene copolymer), when compounded therewith in an amount of say 15% of said rubber, for example.

The nomenclature of the polycarboxylic amino acids referred to herein as starting materials is apparently not yet positively decided. Thus, ethylene diamine N,N'-tetraacetic acid is referred to by Ulrich in U. S. Patent 2,168,181 as tetramethyl-diaminoethane-tetracarboxylic acid. It may also be called N,N'-tetracarboxyl-methyl 1,2-diamino ethane, or ethylene diamine tetracarboxylic acid. This latter name is given in my copending application Serial No. 465,170, filed November 10, 1942, wherein preparation of the esters referred to herein is mentioned, and of which this application is a continuation-in-part. In this connection it should also be pointed out that these polycarboxylic amino acids may be represented by three types of formulas—internal salt, zwitter ion, and carboxylic acid—as is well known in the art of ordinary amino acids. While the carboxylic acid type of formula is shown herein and in the appended claims, it is, therefore, interchangeable with the other two types.

I claim:

1. The tetraisopropyl ester of propylene 1,2-diamine N,N'-tetraacetic acid.

2. The tetrabutyl ester of ethylene diamine N,N'-tetraacetic acid.

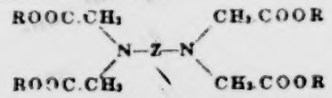
3. The method of preparing an ester of a polycarboxylic diamino acid, which comprises forming an addition product of said diamino acid with sulfuric acid, said addition product containing between about 1.1 and 2 mols of sulfuric acid per mol of said diamino acid, and reacting said addition product with an alcohol, with removal of water of formation, and then neutralizing the sulfuric acid and separating the resulting ester from the other products.



4. The method according to claim 3 wherein said polycarboxylic diamino acid is ethylene diamine N,N'-tetraacetic acid.

5. The method of preparing an ester of a polycarboxylic diamino acid, which comprises forming an addition product of said diamino acid with sulfuric acid by adding an excess of sulfuric acid to said polycarboxylic diamino acid and crystallizing the resulting diamino acid-sulfuric acid addition product in the form of crystals containing between about 1.1 and 2 mols of sulfuric acid per mol of said diamino acid, reacting acid crystals with an alcohol, removing water of formation, and then neutralizing the sulfuric acid and separating the resulting ester from the other products.

6. An ester of a polyacetic amino acid, said ester having the formula



wherein each R is the radical of a lower aliphatic alcohol ROH, and Z is an alkylene group having not less than 2 and not more than 3 carbon atoms.

7. The tetra-ester of ethylene diamine N,N'-tetra-acetic acid with a lower aliphatic alcohol.

8. The tetra-ester of propylene 1,2-diamine N,N'-tetraacetic acid with a lower aliphatic alcohol.

FREDERICK C. BERSWORTH.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Patented Oct. 3, 1950

2,524,218

# UNITED STATES PATENT OFFICE

2,524,218

## WASHING COMPOSITION

Frederick C. Berswo-th, Verona, N. J.

No Drawing. Application March 28, 1946, Serial No. 657,393

3 Claims. (Cl. 252—117)

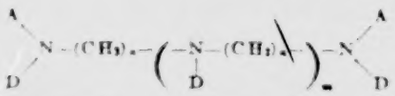
1

This invention relates to derivatives of alkylene polyamines, and more particularly to such compounds which combine the properties of soap compatibility, detergent and wetting action, and ability to sequester metal ions. The synthetic detergents and wetting agents hitherto known fail to combine these properties in the effective manner of the compounds of the present invention.

It is, therefore, an object of this invention to prepare alkylene polyamine derivatives which are detergents and wetting agents, are capable of sequestering metal ions, and are compatible with soap. Other objects will appear in the following disclosure.

It has been shown by Munz, in U. S. Patent 2,240,957, that acetic acid derivatives of amines and diamines are useful for treating hard water. The compounds shown by Munz are not, however, compatible with soap. In my copending application, Serial Number 491,669, filed June 21, 1943, now U. S. Patent 2,407,645, a method of preparing these acetic acid derivatives is shown. Various other methods are known.

The alkylene polyamine derivatives of the present invention are represented by the following formula:



wherein $n$ is either 2 or 3 and $m$ is either zero or a positive integer and wherein D is a member of the group consisting of

$$-CH_2COOH, \quad -CH_2CH_2COOH$$

and their alkali metal salts, and wherein A is a hydrocarbon group containing at least two carbon atoms and having not more than two hydrogen atoms replaced by a member of the group consisting of $-(CH_2)_2COOR$, $-(CH_2)_2COOM$, $-SO_3H$, $-SO_3M$, $-OR$ and $-OR'OR$ wherein R is alkyl or hydrogen, M is an alkali metal, and R' is alkylene, said groups being spaced at least two carbon atoms from the nitrogen atoms.

The compounds of this invention may be prepared in several ways. One of these methods consists in reacting one mol of a suitable polyamine, such as ethylene diamine, trimethylene diamine, diethylene triamine, etc., with two mols of a chloro derivative of the substituent desired in the A positions. The secondary (imino) hydrogens are then substituted either by the chloracetic acid method shown below, or by my process as set forth in my copending application Serial No. 491,669 filed June 21, 1943, now U. S. Patent 2,407,645.

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A second method for preparing these compounds consists of reacting one mol of the alpha beta dichloro derivative of ethylene with two mols of an alpha amine of the substituent desired in the A positions. This compound is then reacted in either of the two general ways described above to provide the proper substituents in the D positions.

It is essential that either two or three ($-CH_2$) groups be present separating the nitrogen atoms and it is further necessary for the purposes of this invention that the groups in the D positions be either acetic acid or propionic acid radicals or their alkali metal salts. These conditions insure sequestering action of the compound and tend to make the compound water miscible. The presence of more or less than the specified number of ($-CH_2$) groups seriously reduces or eliminates this sequestering action.

It has been discovered that in general the longer the hydrocarbon chain in the A positions the more soap-like the entire compound becomes. It has been further found that when this chain is four carbon units in length or longer it is water-dispersible, and when it is more than seven carbon units in length the calcium complex is soluble in water, foams strongly, cleans well and is compatible with soap.

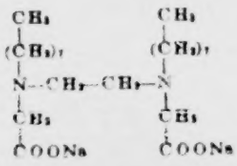
Another requirement of these compounds is that any hydrophyllic groups attached to the hydrocarbon chain must be at least two carbon atoms removed from the nitrogen atoms. The substitution of a relatively long alkyl chain in the primary nitrogen position also destroys any electrolyte effect of the compound. It becomes possible, therefore, with proper substitutions in the A positions to prepare compounds which are miscible with materials such as soap without causing any deleterious effects to the soap, and form emulsions with liquid hydrocarbons, and other water insoluble organic compounds.

In order to understand this invention more clearly the following examples are given:

*Example 1*

One mol of ethylene diamine added to two mols of octyl chloride is heated under atmospheric pressure until the reaction is substantially complete. The resulting compound is reacted with chloracetic acid in methyl alcohol keeping the reaction solution at a pH of 8 to 8.5. The mixture is heated and held to a gentle refluxing of the methyl alcohol for a period of six to eight hours. The material is filtered from the formed sodium chloride and the methyl alcohol is distilled off. The product is a semi-solid paste with an appearance somewhat like stearic acid and is soluble

in alkalies and acids, compatible with soap and has the power of sequestering metal ions. It is believed that this compound has the following formula:



*Example 2*

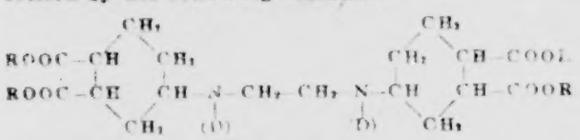
Two mols of hexyl amine are heated with symmetrically substituted trimethylene dichloride in methyl alcohol. The resulting product is treated according to my application Serial Number 491,669 filed June 21, 1943, now U. S. Patent 2,407,645. This method, briefly, consists of adding two mols of formaldehyde and two mols of sodium cyanide slowly and in small balanced quantities under strongly alkaline conditions to the diamine and heating the mixture with rapid agitation. The compound is compatible with soap and is capable of sequestering metal ions such as calcium ions.

Acrylic type unsaturated acids may be used advantageously in the preparation of compounds included within the scope of this invention. Several methods of employing those unsaturated acids are possible. In general they are first esterified to form a stable ester. Two mols of the ester may then be reacted directly with a diamine, such as ethylene diamine, according to the following equation:

$$NH_2RNH_2 + 2CH_2{=}CH(CH_2)_nCOOR' \rightarrow R'OOCCH_2(CH_2)_{n+1}HNRNH(CH_2)_{n+1}CH_2COOR'$$

It is noted that the ester group is sufficiently stable so that the hydrogen of the primary amino group acts as a hydrogen donor saturating the double bond. The remaining hydrogen of the amino groups is then reacted according to either of the methods described above. R in the case just described may be $-(CH_2)_n-(NH(CH_2)_n)_m-$ where $n$ is 2 or 3 and $m$ is 0, 1 or 2. A second general method of utilizing an unsaturated ester is hydro-halogenation in the presence of methyl alcohol using dry HCl to form the chloride ester. The halogen ester is then reacted with a polyamine in methyl alcohol in the ratio of two mols of ester to one of the polyamine as in the previous cases. It will be noted that only the primary hydrogens are treated as being in the A positions.

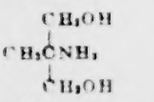
In addition to the compounds described above derivatives using the condensation products from the Diels-Alder reaction may be used. A preferred compound of this series is produced from maleic anhydride and butadiene and is represented by the following formula:



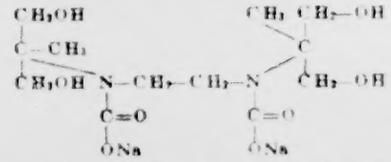
The dicarboxylic acid resultin, from the condensation of the butadiene and maleic anhyuride is condensed with ethylene diamine in the same manner that the unsaturated acids described above are condensed, that is, the di-acid is esterified and is reacted with hydrochloric acid and the hydrochloride is in turn reacted with ethylene diamine or other suitable diamines, triamines or tetramines. As in the other compounds described, the secondary hydrogens attached to the nitrogen groups are reacted to substitute acetic acid or propionic acid radicals. The product is compatible with soap and is an excellent cleaning agent. If desired, the ester may be saponified to yield the alkali metal salt, and if the acid is preferred, the salt is treated with an acid in the conventional manner.



Still other types of compounds may be produced by reacting 2 amino, 2 methyl, 1-3 propane diol, the formula of which is



with ethylene dichloride. The compound 2 amino, 2 methyl, 1-3 propane diol is dispersed in methyl alcohol. To this well dispersed, well agitated suspension two mols of ethylene dichloride are added to each ten mols of the amine. The mixture is refluxed for about 1 hour. Sodium carbonate is added from time to time to maintain the pH between 8.0 and 8.5. The reaction is over in approximately 5 hours and requires about two mols of sodium carbonate and 1500 cc. of methyl alcohol. Sodium chloride formed is removed by filtration and the alcohol is removed by distillation. Secondary hydrogen atoms on each of the nitrogen atoms are reacted according to my application Serial Number 491,669 filed June 21, 1943, now U. S. Patent 2,407,645 and are substituted by the radical $CH_2COONa$. The formula of the resulting product is believed to be as follows:



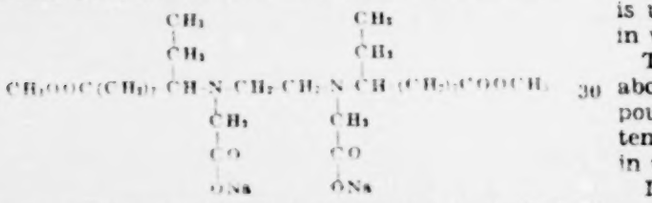
The product as produced is a yellow-white solid. It has been discovered that if the amine diol is reacted according to the chloracetic acid method some of the hydroxyl groups are involved to form ether acids. The compound forms a clear calcium complex and is an excellent detergent. While it increases the foam of soap solutions it, by itself, is not a foaming agent as are the other compounds described above.

A preferred type of compound of this invention is the derivative of ethylene diamine in which a long chain acid such as undecylenic acid is substituted in the A positions. Two compounds using undecylenic acid are of particular interest and the method of making them is described below.

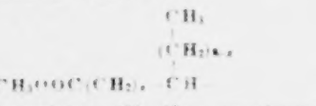
The methyl ester of the acid is first produced. This is in turn reacted with hydrobromic acid. This halogenation is carried out in ligroin in the presence of air and at a temperature of 0° C. Two mols of the bromide ester react readily with ethylene diamine in the presence of methyl alcohol as a solvent. The reaction is carried out at a pH of 8.0 which is regulated by the addition of dry caustic soda. Sodium bromide is precipitated and may be filtered off. When 70% ethylene diamine is used some foaming will be experienced due to the water present. It has been found that the best procedure is to dehydrate the entire reaction mixture, after the reaction is completed, with benzene. There is no appreciable hydrolysis of the methyl ester during this reaction. After removing the alcohol by distillation, a compound is obtained of a yellowish

semi-crystalline mass, soluble in organic solvents and miscible with water to form a milky solution. To react the secondary hydrogen atoms on the nitrogen atoms, the product produced above is redissolved in methyl alcohol and the alcohol solution added to a slurry of chloracetic acid sodium salt in methyl alcohol. Two mols of such salts are used per mol equivalent of the reaction product. There is considerable heat of reaction but if this is permitted to run its course using 200 cc. of methyl alcohol for each mol of chloracetic acid sodium salt, the heat of reaction is 60° C. After two hours sodium bromide begins to precipitate and heat is applied to maintain the reflux temperature of methyl alcohol for about six to eight hours. Sodium carbonate is added from time to time to regulate the pH between 7.5 and 8.5. Provision must be made to allow liberated carbon dioxide to escape. When the reaction is completed the water that is present due to the hydrous diamine is removed before filtering.

When the undecylenic acid used in the preparation of the foregoing product is of the usual type, $CH_3—CH_2—CH_2—(CH_2)_7—COOH$ (9-hendecenoic acid), the formula of that product is as follows:



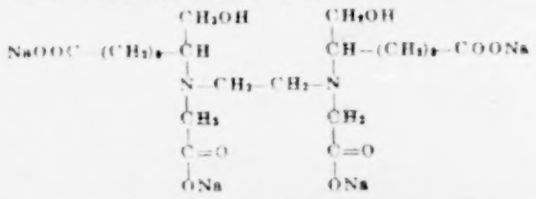
It is obvious that the lengths of the two alkyl chains on either side of the carbon atom of the undecylenic acid ester radical attached to the nitrogen atoms will depend upon the location of the double bond in the undecylenic acid, as well as upon which carbon atom the halogen atom is attached. Hence a more general formula for these compounds, using the methyl ester of any undecylenic acid, will be represented by the following radical in the A positions:



wherein, conformably with the requirements of the compounds of this invention, $x$ is not less than 3—and is, of course, not greater than 8.

The above product is illustrative of these highly effective compounds. They act as cleaners and foam in any type of water. They are not precipitated or salted out by any amount of electrolyte and are powerful wetting agents even in water of 400 parts per million hardness and in sea water. They are compatible with soap and are not precipitated in the form of lime soap nor is the soap salted out. They have a unique dual action in that they will form highly soluble filtrable calcium complexes. They also have the property of dispersing lime salts in an amount far greater than that calculated to form the clear complex. The calcium complex itself is soluble in alcohol. It may be noted that the compounds of this invention are both anionic and cationic in nature. For these reasons, and because these compounds are extremely stable in both alkaline and acid solutions, they have wide usefulness.

A second, and slightly different type of compound from that described above may be prepared from omega undecylenic acid by reacting the acid ester with hypochlorous acid, HOCl. This acid acts much the same way as does hydrochloric acid in its addition to the double bond of undecylenic acid. The addition product is reacted with ethylene diamine and the secondary hydrogen atoms are reacted as described above; upon hydrolysis of the ester groups with alkali a compound of the following formula results:

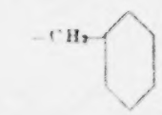


This compound is very similar in nature to the compound described above except that it is more soluble in water. Likewise, the lengths of the two alkyl chains of the undecylenic acid radical will depend upon conditions as set forth above.

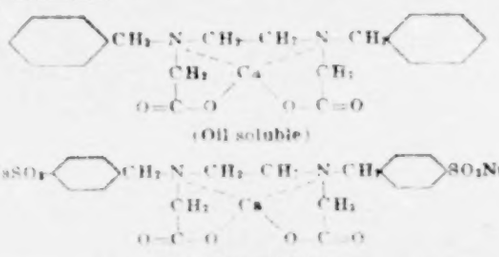
When the hydrochloric acid derivative of undecylenic acid ester is reacted with diethylene triamine, and the secondary hydrogen atoms on the three nitrogen atoms are reacted with chlorpropionic acid sodium salt, there is produced a compound similar to the ethylene diamine derivative wherein the chloracetic acid sodium salt is used. The product is, however, more soluble in water.

The resulting esters formed by the reactions above may be hydrolyzed (as shown in the compound above) or may be retained. Ester groups tend to decrease the solubility of the compound in water.

If the A groups are insoluble or immiscible in water, the calcium complex of the compound is not soluble in water and precipitates. For instance if A is



the calcium complex precipitates in water. However, if this benzene nucleus is sulfonated the sodium sulfonate calcium complex is soluble and is useful for the purpose of this invention. If not sulfonated the compound is quite oil soluble, while the sulfonated compound (sodium salt) is quite water soluble. The compounds described above may be illustrated by the following formula:



(Oil soluble)

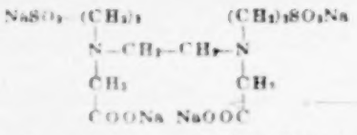


(Water soluble)

This compound illustrates the fact that materials particularly suitable for cleaning action and sequestering ability in oil may be secured, by substituting suitable oil soluble groups in the A positions.

The introduction of a $(—SO_3Na)$ group into the compounds of this invention may be readily accomplished by reacting two mols of a compound such as taurine, $NH_2(CH_2)_2SO_3H$, with each one mol of dichloro ethylene, $Cl(CH_2)_2Cl$. The reaction follows the general procedure outlined for that described of reacting 3 amino, 2 methyl, 1-3 propane diol with ethylene dichloride de-

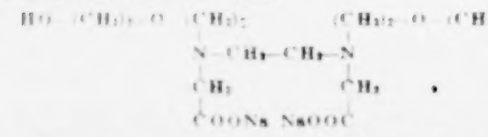
scribed above. The resulting product is then reacted according to my application Serial Number 491,669, now U. S. Patent 2,407,645, to substitute the secondary hydrogen atoms on the nitrogen atoms with (—$CH_2COONa$) groups. The completed compound is represented by the following formula:



The product may be crystallized from methyl alcohol solutions.

It is excellent as a detergent and for sequestering metal ions, and is completely miscible with soap.

Other types of radicals in the A positions are also advantageous for some cleaning purposes; for example, compounds prepared by reacting an hydroxylated ether with a diamine. The reaction between diethylene glycol and anhydrous ethylene diamine may be carried out merely by boiling the two together. Xylene or a dehydration catalyst may be added but such precaution is not essential. As in the previous examples, the secondary hydrogen atoms on the nitrogen atoms are substituted with —$CH_2COONa$ or —$CH_2CH_2COONa$ groups, and by the same method. The final compound may be represented by the formula:



This compound is an excellent detergent and sequestering agent and is completely miscible with soap.

A summary of the properties of the compounds herein described will serve to point out obvious uses for them.

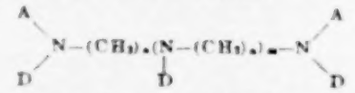
As prepared, the compounds have no deleterious electrolytic properties to soap. Accordingly as they are compatible with soap when incorporated with soap in which electrolytes are present, the electrolytes do not adversely affect the soap. They are of themselves good wetting agents and detergents but do not defat the skin. Being completely stable in both alkali and acid solutions, they may be incorporated in media of either type. The effectiveness of these compounds may be due, in part, to the fact that they are both cationic and anionic in action as well as the fact that they sequester metal ions, such as calcium ions, thus preventing the precipitation of calcium soaps. It can be seen from the above description that the compounds may be varied to make them soluble in oils, organic solvents, or water. The foaming power of the compound may be adjusted, as the longer the aliphatic chain in the A positions, the greater the foaming power, while the heterocylic compound shown has very little foaming action (although it is a good detergent). More than six carbon atoms in the hydrocarbon chain insures the compound of having good wetting action, particularly if carboxyl groups or hydroxyl groups are present.

In the above discussion of this invention wherein alkali metal salts are disclosed, and in the claims, the term "alkali metal" is intended to include the group consisting of sodium, potassium, and ammonium (—$NH_4$).

Although any desired amount of these compounds may be added to a water-soluble soap to form a washing compound, it has been found that from 1% to 5% by weight prevents the precipitation of calcium or magnesium soaps in any commonly used water, and yields a good detergent soap.

I claim:

1. A detergent composition consisting of a water soluble alkali metal-fatty acid soap and an alkylene polyamine derivative, the amount of said alkylene polyamine derivative in the said mixture being from 1% to 5% by weight of the mixture, said alkylene polyamine derivative being one conforming to the formula:

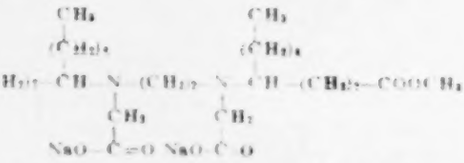


wherein $n$ is an integer selected from the group consisting of 2 and 3; $m$ has a value equal to one of the group consisting of zero, the numeral one (1), and any multiple of the numeral one (1); D is a member of the group consisting of —$CH_2COOH$, —$CH_2CH_2COOH$, and their alkali metal salts; A is a hydrocarbon group containing at least two carbon atoms and having not more than two hydrogen atoms replaced by a member of the group consisting of

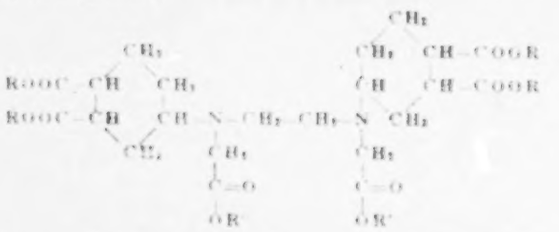
—$(CH_2)_xCOOR$, —$(CH_2)_xCOOM$,
—$SO_3H$, —$SO_3M$, —OR

and OR'OR wherein R is one of the group consisting of alkyl and hydrogen, M is an alkali metal, and R' is an alkylene group, said replacing groups being spaced from the N atoms of the compound by at least two carbon atoms of the hydrocarbon group containing the same.

2. The detergent composition of claim 1, wherein said alkylene polyamine derivative comprises the compound:



3. The detergent composition of claim 1, wherein said alkylene polyamine derivative comprises the compound:



wherein R is one of the group consisting of hydrogen and alkyl and R' is one of the group consisting of hydrogen and alkali metal.

FREDERICK C. BERSWORTH.

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| 2,407,645 | Bersworth | Sept. 17, 1946 |
| 2,412,945 | Bersworth | Dec. 24, 1946 |

Patented Sept. 17, 1946 2,407,645

# UNITED STATES PATENT OFFICE

2,407,645

## ALIPHATIC POLYCARBOXYLIC AMINO ACIDS AND PROCESS OF MAKING THEM

Frederick C. Bersworth, Verona, N. J., assignor to The Martin Dennis Company, Newark, N. J., a corporation of New Jersey

No Drawing. Application June 21, 1943, Serial No. 491,669

10 Claims. (Cl. 260—534)

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This invention relates to polycarboxylic amino acids, and more particularly to a process for producing aliphatic poly-carboxylic amino acids and alkali metal salts thereof.

In the past, attempts have been made to make aliphatic polycarboxylic amino acids but they have all been characterized by relatively low yields or high cost and, consequently, have been of limited commercial value. The most successful of these previous methods is the choloracetate reaction as shown by Munz in U. S. Patent 2,130,595. The reaction between formaldehyde, a cyanide, and an amine to form an amino acid is known, and has been used with aromatic amines and with ammonia; however, as far as I am aware that type of reaction has never been used satisfactorily with aliphatic amines.

It is an object of this invention to produce aliphatic polycarboxylic amino acids by a process whereby the yields are satisfactory. Another object is to produce these amino acids using an alkali cyanide, formaldehyde, and a primary or secondary aliphatic amine by a process whereby the reaction goes smoothly and is a substantial improvement over all prior methods. Another object is to show a procedure for making aliphatic polycarboxylic amino acids on a commercial scale. Still another object is to produce the alkali metal salts of these amino acids.

Briefly, these objects may be accomplished by preparing and maintaining a solution in which free unreacted aliphatic amine and alkali metal cyanide are both always present, and with the amine in excess over the cyanide, at least until the completion of the reaction, and adding slowly to said solution about one mol of formaldehyde for each amino-hydrogen of the amine to be replaced so that ammonia formed will be as completely removed as possible before more formaldehyde is added, and add about the same total molal quantity of an alkali metal cyanide at such a rate that the cyanide is maintained in excess over the formaldehyde. The solution is constantly agitated to prevent any local undesirable excess concentrations of the reactants and the mixture is heated to a suitable temperature. The alkali metal salt of the polycarboxylic amino acid is formed together with by-products. The free acid may be precipitated from this crude reaction product by adding any strong acid in

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proper quantity. The pure alkali metal salt may then be obtained by dissolving the acid in a solution of the proper hydroxide. Alternatively, it may be obtained by crystallization from the crude reaction product.

As this invention is based primarily upon the particular method of operation, an explanation of the chemistry of the reaction and therefore the reasons for the method used is given.

In the reaction mixture at any one time during, and prior to completion of, the reaction are present the compounds introduced, which include the alkali metal hydroxide, the amine, the alkali metal cyanide and formaldehyde together with products formed by the reaction, which products include ammonia. The object to be obtained is the promotion of the inter-reaction between the formaldehyde, the cyanide, and the amine while preventing the side reactions with ammonia or reaction of formaldehyde and the amine alone or formaldehyde and the cyanide alone. The rate of reaction of each of the possible combinations determines the method and conditions for carrying out the invention.

The sodium hydroxide does not enter into the reaction and therefore all of it may be introduced in the beginning. Its purpose is to provide a sufficiently alkaline reaction medium (if the other ingredients do not do so) to prevent hydrolysis of the cyanide with consequent formation of HCN, the presence of which is highly undesirable, because of both reactivity and toxicity. All the amine may also be introduced into the reaction vessel at the start because it does not react with either the cyanide or ammonia alone. Formaldehyde, however, reacts violently with the cyanide and rapidly with the amine and must be added in such a manner that as little free formaldehyde is present at any given time as possible, consistent with practical operating procedure. As the rate of reaction of formaldehyde is greater with the cyanide than with the amine, the concentration of the cyanide, in the presence of formaldehyde, must not be allowed to become too high or the formaldehyde will react with the cyanide alone to form glycolates, materially reducing the yield of desired product. It has therefore been found that no more than about 25% of the total cyanide necessary to react with all the replaceable hydrogen atoms attached to the amino nitrogen atoms

of the amine (in the presence of formaldehyde) should be added, or present as free cyanide in the reaction mixture at any one time; smaller more frequent additions are preferable, and may amount to a continuous slow addition of the cyanide. But at all times the cyanide should be in excess of the formaldehyde and preferably by at least about 5% (calculated on the basis of total mols of formaldehyde or cyanide necessary to react with all the replaceable hydrogen atoms attached to the amino nitrogen atoms of the amine), in order that the amine, cyanide, and formaldehyde will react properly and without excessive formation of by-products. It has been further found that the yields of finished product become better as the concentration of free formaldehyde in the reaction mixtures is reduced. However, economic factors enter so that the yield obtained in practical operation is balanced against the time necessary for the reaction.

Ammonia which is formed by the reaction between the amine, cyanide, and formaldehyde must be removed from solution at once because if present in appreciable concentration, it will react with formaldehyde to form hexamethylenetetramine, a by-product which, because it lowers the yield, is undesirable.

The temperature at which the reaction takes place is also of importance; if it is too low, the reaction goes too slowly and the liberation of ammonia is not sufficiently complete; but if too high, colored polymers of cyanides form. The temperature of boiling is, of course, a function of the pressure; at reduced pressure the liquid may be boiled and the ammonia removed at a temperature below that at which the colored by-products form. It has been found that at about a ten-inch vacuum the reaction goes smoothly in the range of 60°–80° C. When the reaction is substantially completed some free alkali cyanide is still present; this may be reacted by introducing additional formaldehyde and operating at atmospheric pressure or somewhat higher, and raising the temperature to the boiling point (110–120° C. or somewhat higher). This additional formaldehyde (about 10% of the theoretical total formaldehyde is usually needed) should be added to replace the formaldehyde lost through by-product formation and by other means, and to make up the approximately 5% deficiency in formaldehyde (below cyanide) already referred to. This additional formaldehyde serves to increase the yield and to eliminate all traces of cyanide; the reaction product should be free from all cyanide when completed, at least for most commercial uses.

To avoid any local excess concentration, and to maintain a constant temperature through the mixture, it is desirable that it be constantly agitated.

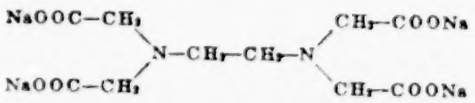
Although the exact mechanism of the reaction is not known, it is believed that a nitrile is first formed by the reaction of the alkali cyanide with formaldehyde and the amine. The nitrile is immediately hydrolyzed and converted to the final sodium salt of the aliphatic amino carboxylic acid, also giving ammonia as a by-product. It has been found that if an alkali metal hydroxide is added to the mixture the hydrolysis of the alkali metal cyanide is repressed, thus preventing the formation of complex cyanide derivatives, and has the additional advantage of aiding the rapid and complete hydrolysis of the nitrile formed, which is desirable. The pH of the reaction mixture must be at least about 9 and preferably higher.

If the above sequence is not performed, the reaction may go with explosive violence and no appreciable yield of the desired product is formed, or the reaction will give many undesirable by-products.

To further illustrate the process of the invention the following examples are given. It is to be understood that these examples are illustrative rather than limiting.

### *Example I*

10 mols of ethylene diamine as a 30% aqueous solution and 4 mols of solid caustic soda are placed in a steam heated kettle supplied with an agitator. Eight mols of sodium cyanide as a concentrated water solution (about 30%) are added and the solution heated to 60° C. About a 10 inch vacuum is applied to bring the liquid to incipient boiling. Formaldehyde (7.5 mols of 37–40% aqueous solution) is slowly added, the temperature being held at 60° C., and the solution vigorously stirred. Then, when the evolution of ammonia has substantially stopped, eight more mols of sodium cyanide, followed by eight mols of formaldehyde are added as before. This is continued until 40 mols of cyanide and forty mols of formaldehyde have been added. Then at the end about 2 mols more of formaldehyde are added, making forty-two in all, to remove any last traces of cyanide. About 8 to 10 hours are required to complete the reaction. The resulting product, referred to herein as the crude reaction product, is essentially an aqueous solution of the sodium salt of ethylene diamine tetraacetic acid.



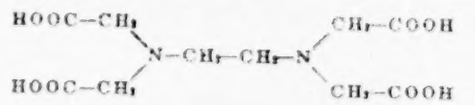
together with various by-products. On acidifying to a pH between .75 and 2, the corresponding free acid precipitates, and has been identified by ultimate analysis and electrometric titration curves.

### *Example II*

Using the procedure of Example I, but with the following total quantities, a still better yield was obtained:

| | Pounds |
|---|---|
| Ethylene diamine (70%) | 30 |
| NaOH (anhydrous) in 16 lbs. $H_2O$ | 8 |
| Formalin (37%) | 137 |
| NaCN (96%) in 240 lbs. $H_2O$ | 80 |

When the reaction was completed, 500 lbs. of solution were obtained which, when acidified with HCl (about 165 lbs. of 38% HCl, or a pH of about 1.5), produced a yield of 81.6% of theoretical of



based upon the weight of ethylene diamine used.

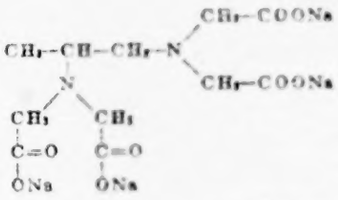
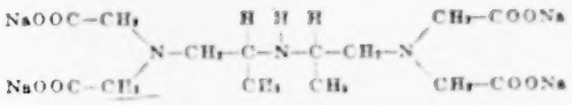
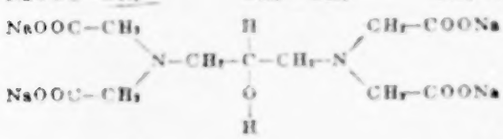
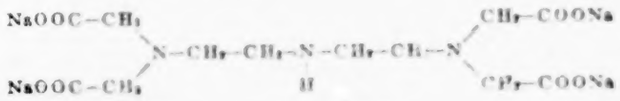
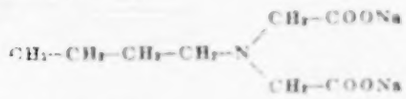
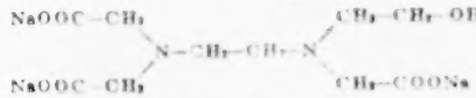
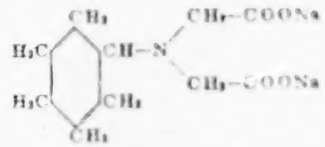
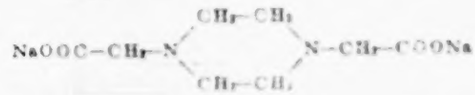
I have found that any alkali metal cyanide or any alkali metal hydroxide may be substituted for the sodium cyanide and sodium hydroxide, respectively, used in the above examples; an alkali hydroxide is not essential though desirable, as already pointed out.

Any substance yielding formaldehyde under the conditions of this process, e. g. paraformalde-

hyde, may be used in place of formaldehyde in carrying out the process of this invention.

In place of ethylene diamine any aliphatic amine having at least two replaceable hydrogen atoms attached directly to the same or different amino nitrogen atoms may be used to produce the polyacetic acid aliphatic amino compounds described herein, and therefore such amines are equivalent to ethylene diamine, as illustrated in more detail below.

A few examples of various amines that have been reacted with an alkali metal cyanide and formaldehyde by the procedure of the foregoing examples, and the products obtained therefrom, are given below.

| *Amine* | *Product* |
|---|---|
| 1-2 propylene diamine |  |
| Dipropylene triamine |  |
| 1-3 diamino propanol-2 |  |
| Diethylene triamine |  |
| N-butylamine |  |
| N-monoethoxy ethylene diamine |  |
| Cyclohexyl amine |  |
| Piperazine |  |

When polyamines having more than two amino nitrogen atoms are used, e. g. dipropylene triamine and diethylene triamine as shown in the foregoing tabulation, the hydrogen atoms on the intermediate amino nitrogen atoms as well as those on the terminal amino nitrogen atoms may be replaced by —$CH_2COOM$ groups (M being an alkali metal), by carrying still further the reaction with formaldehyde and alkali metal cyanide. In so doing, the precautions already pointed out as to relative amounts of reactants must be observed; furthermore, since these higher-than-tetra substitutions are less readily made than those of tetra or lower, the reaction is preferably carried out at or above atmospheric pressure, and at or near the boiling point at the pressure used. The resulting penta- (and higher) alkali metal penta- (or higher) acetic acid salts cannot be made by previously described processes such as the chloracetic acid process, so far as I am aware, and these salts and their corresponding acids are, I believe, new compounds.

As shown by the formulae given above, the sodium (or other alkali-metal) salt of the amino acid is formed in each case. These salts are extremely soluble, and in order to purify them, advantage is taken of the extreme insolubility of the corresponding acids. The solution is acidified to a pH of .75 to 2 with an acid and the pure amino acid separates out in the form of white crystals.

When a diamine containing two replaceable hydrogen atoms attached to each amino nitrogen atom is used as the amine to produce a tetra-substituted product, it may be desirable to make alkali metal salts in which fewer than four of the hydrogen atoms of the four —COOH groups are replaced by the alkali metal. Procedure for doing so is described below.

The tetra-alkali-metal salt may be prepared by merely adding sufficient solution of an alkali metal hydroxide to replace all the acid hydrogens of the free acid. The tri-alkali-metal salt is unstable except perhaps in solution and apparently cannot be isolated. The di-alkali salt is the least soluble of the alkali salts, and may be readily crystallized from solution. The following is an example of the preparation of the disodium salt of ethylene diamine tetraacetic acid. To 1000 gms. of the crude reaction product as obtained in accordance with Example 1, 264 gms. of ethylene diamine tetracetic acid are added. The mixture is preferably heated to incipient boil-

ing to increase the rate of reaction, and then the mixture is allowed to cool and crystallize. The crystals formed are filtered off, washed with the smallest possible amount of ice water, and dried to a constant weight, which is 452 gms. A representative sample of the product so prepared showed, upon analysis, 13.26% sodium against a theoretical of 13.70% for the disodium salt. The dialkali salt has a pH of about 5.3 and behaves like a weak acid, displacing $CO_2$ from carbonates and reacting with metals to form hydrogen. It is a white crystalline solid.

The monoalkali-metal salt may be prepared by heating together equal molecular quantities of the dialkali salt and the free acid in the presence of water. When the solution is cold, only the dialkali dissolves, but upon heating both go into solution. The mono salt may be recovered by evaporating the solution to dryness. If the above solution is cooled, the free acid precipitates out thus showing that the mono salt is unusual in that it is more stable at high temperature than at low temperature.

The chemical properties of these polycarboxylic amino-acids and their alkali salts make them valuable for many purposes. It is well known that the alkali salts are useful in water-softening and, when mixed with soap, as washing agents. These amino-acids also serve as starting materials for useful products such as amino polycarboxylic acid esters as shown in my application Serial No. 491,670 filed June 21, 1943.

This is a continuation-in-part of my copending application Serial No. 400,967, filed July 3, 1941.

I claim:

1. The process of preparing aliphatic polycarboxylic amino acids which comprises reacting an aliphatic amine having at least one replaceable hydrogen atom attached directly to an amino nitrogen atom, with an alkali metal cyanide and a formaldehyde-yielding substance under conditions sufficiently alkaline that there is substantially no hydrolysis of the said cyanide and in total amounts such that for each such replaceable hydrogen atom one molecule of said cyanide and one molecule of said formaldehyde-yielding substance are added during the reaction, while maintaining the amount of said free cyanide present in the reaction mixture at any one time at not more than about one quarter of the amount necessary to react completely with the original amine, and maintaining the amount of free formaldehyde-yielding substance present at any one time at less than one mol thereof per mol of said free cyanide present until substantial completion of the reaction of said amine has been effected, and removing ammonia from the reaction mixture so that substantially no free ammonia is present therein at any time during the reaction, and finally, after all the cyanide has been added, adding at least sufficient formaldehyde-yielding substance to react with any unreacted cyanide.

2. The process according to claim 1 wherein the reaction is carried out at a temperature of between about 60° C. and about 80° C. and at a pressure such that the reaction mixture is substantially at the boiling point at said temperature, until the major part of the reaction has been completed, and the temperature and pressure are then gradually raised while maintaining susbtantially boiling conditions, until completion of the reaction.

3. The process of preparing aliphatic polycarboxylic amino acids which comprises reacting an aliphatic amine having at least one replaceable hydrogen atom attached directly to an amino nitrogen atom, with an alkali metal cyanide and a formaldehyde-yielding substance under conditions sufficiently alkaline that there is substantially no hydrolysis of the said cyanide and in total amounts such that for each such replaceable hydrogen atom one molecule of said cyanide and one molecule of said formaldehyde-yielding substance are added during the reaction, while maintaining the amount of said free cyanide present in the reaction mixture at any one time at not more than about one quarter of the amount necessary to react completely with the original amine, and maintaining the amount of free formaldehyde-yielding substance present at any one time at less than one mol thereof per mol of said free cyanide present until substantial completion of the reaction of said amine has been effected, and removing ammonia from the reaction mixture so that substantially no free ammonia is present therein at any time during the reaction, and finally, after all of the cyanide has been added, adding a slight excess of formaldehyde-yielding substance over that necessary to react with any unreacted cyanide.

4. The process according to claim 1 wherein the formaldehyde-yielding substance is formaldehyde.

5. The process according to claim 1 wherein the amine is a primary aliphatic amine.

6. The process according to claim 1 wherein the amine is a primary aliphatic diamine.

7. The process of preparing the sodium salts of ethylene diamine tetraacetic acid which comprises reacting ethylene diamine with sodium cyanide and formaldehyde in the presence of sufficient sodium hydroxide to bring the pH to at least about 9 while maintaining a temperature of between about 60° C. and about 80° C. and a pressure such that the reaction mixture is substantially at the boiling point at said temperature, and maintaining the amount of free cyanide present in the reaction mixture at any one time at not more than one quarter of the amount necessary to react completely with the ethylene diamine, and maintaining the amount of free formaldehyde present in the reaction mixture at any one time at about five per cent less than the amount of sodium cyanide present, which five percent is based on the total quantity of sodium cyanide to be reacted, said sodium cyanide being added until four mols thereof have been added for each mol of ethylene diamine and the formaldehyde being added until at least about 95 per cent of the theoretically required four mols have been added, adding sufficient additional formaldehyde substantially completely to react with the unreacted sodium cyanide, increasing the pressure and heating the solution to boiling at a temperature at least about 110–120° C., and removing ammonia from the reaction mixture so that substantially no free ammonia is present therein at any time.

8. The process according to claim 1 wherein the amine is diethylene triamine.

9. The process according to claim 1 wherein the amine is n-butylamine.

10. The process of preparing aliphatic polycarboxylic amino acids which comprises reacting an aliphatic amine having at least one replaceable hydrogen atom attached directly to an amino nitrogen atom, with an alkali metal cyanide and a formaldehyde-yielding substance under conditions sufficiently alkaline that there is substantially no hydrolysis of the said cyanide,

while maintaining the amount of said free cyanide present in the reaction mixture at not more than about one quarter of the amount necessary to react completely with the original amine, and maintaining the amount of free formaldehyde-yielding substance present at less than one mol thereof per mol of said free cyanide present and continuing to add said cyanide and said formaldehyde-yielding substance under said conditions until each said replaceable hydrogen atom has been replaced by the radical $-CH_2COOM$ wherein M is an alkali metal, and removing ammonia from the reaction mixture so that substantially no free amomnia is present therein at any time during the reaction, and finally, after all the cyanide has been added, adding a slight excess of formaldehyde-yielding substance over that necessary to react with any unreacted cyanide.



FREDERICK C. BERSWORTH.

Patented Dec. 5, 1950

2,532,391

# UNITED STATES PATENT OFFICE

2,532,391

## ALKYLENE POLYAMINE DERIVATIVES

Frederick C. Bersworth, Verona, N. J.

No Drawing. Application March 15, 1947, Serial No. 735,018

3 Claims. (Cl. 260—404.5)

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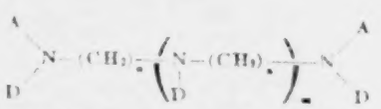
This invention relates to derivatives of alkylene polyamines, and more particularly to such compounds which combine the properties of soap compatibility, detergent and wetting action, and ability to sequester metal ions. The synthetic detergents and wetting agents hitherto known fail to combine these properties in the effective manner of the compounds of the present invention.

It is, therefore, an object of this invention to prepare alkylene polyamine derivatives which are detergents and wetting agents, are capable of sequestering metal ions, and are compatible with soap. Other objects will appear in the following disclosure.

It has been shown by Munz, in U. S. Patent 2,240,957, that acetic acid derivatives of amines and diamines are useful for treating hard water. The compounds shown by Munz are not, however, compatible with soap in more than very small amounts. In my U. S. Patent 2,407,645, filed June 21, 1943, a method of preparing these acetic acid derivatives is shown. Various other methods are known.

The alkylene polyamine derivatives of the present invention are represented by the following formula:



wherein $n$ is either 2 or 3 and $m$ is either zero or a positive integer and wherein D is a member of the group consisting of —$CH_2COOH$, —$CH_2CH_2$-COOH, and their alkali metal salts, and wherein A is a hydrocarbon group containing at least two carbon atoms and having not more than two hydrogen atoms replaced by a member of the group consisting of —$(CH_2)_2COOR$, —$(CH_2)_2$-COOM, —$SO_3H$, —$SO_3M$, —OR and —OR'OR wherein R is alkyl or hydrogen, M is an alkali metal, and R' is alkylene, said groups being spaced at least two carbon atoms from the nitrogen atoms. The term alkali metal as used above, and in the claims, is intended to include particularly sodium, potassium, and ammonium.

The compounds of this invention may be prepared in several ways. One of these methods consists in reacting one mol of a suitable polyamine, such as ethylene diamine, trimethylene diamine, diethylene triamine, etc., with two mols of a chloro derivative of the substituent desired in the A positions. The secondary (imino) hydrogens are then substituted either by the chloracetic acid method shown below, or by my process

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as set forth in my U. S. Patent 2,407,645 filed June 21, 1943.

A second method for preparing these compounds consists of reacting one mol of the alpha beta dichloro derivative of ethylene with two mols of an alpha amine of the substituent desired in the A positions. This compound is then reacted in either of the two general ways described above to provide the proper substituents in the D positions.

It is essential that either two or three (—$CH_2$) groups be present separating the nitrogen atoms and it is further necessary for the purposes of this invention that the groups in the D positions be either acetic acid or propionic acid radicals or their alkali metal salts. These conditions insure sequestering action of the compound and tend to make the compound water miscible. The presence of more or less than the specified number of (—$CH_2$) groups seriously reduces or eliminates this sequestering action.

It has been discovered that in general the longer the hydrocarbon chain in the A positions the more soap-like the entire compound becomes. It has been further found that when this chain is four carbon units in length or longer it is water dispersible, and when it is more than seven carbon units in length the calcium complex is soluble in water, foams strongly, cleans well and is compatible with soap.

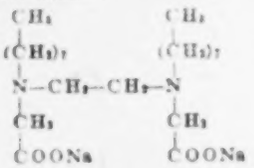
Another requirement of these compounds is that any hydrophyllic groups attached to the hydrocarbon chain must be at least two carbon atoms removed from the nitrogen atoms. The substitution of a relatively long alkyl chain in the primary nitrogen position also destroys any electrolyte effect of the compound. It becomes possible, therefore, with proper substitutions in the A positions to prepare compounds which are miscible with materials such as soap without causing any deleterious effects to the soap, and form emulsions with liquid hydrocarbons, and other water insoluble organic compounds.

In order to understand this invention more clearly the following examples are given:

### *Example 1*

One mol of ethylene diamine added to two mols of octyl chloride is heated under atmospheric pressure until the reaction is substantially complete. The resulting compound is reacted with chloracetic acid in methyl alcohol keeping the reaction solution at a pH of 8 to 8.5 by the gradual addition of sodium hydroxide. The mixture is heated and held to a gentle re-

fluxing of the methyl alcohol for a period of six to eight hours. The material is filtered from the formed sodium chloride and the methyl alcohol is distilled off. The product is a semi-solid paste with an appearance somewhat like stearic acid and is soluble in alkalies and acids, compatible with soap and has the power of sequestering metal ions. It is believed that this compound is the sodium salt of N,N' di-octyl ethylene diamine N,N' di-acetic acid having the following formula:



The sodium, potassium, and ammonium salts of the above compound and those shown in the subsequent examples may be prepared by conventional methods from the corresponding acids.

*Example 2*

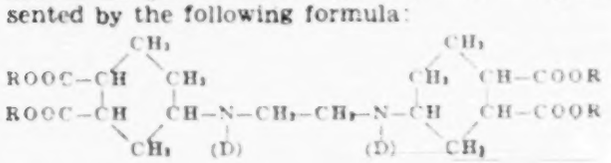
Two mols of hexyl amine are heated with symmetrically substituted trimethylene dichloride in methyl alcohol. The resulting product is treated according to my U. S. Patent 2,407,645 filed June 21, 1943. This method, briefly, consists of adding two mols of formaldehyde and two mols of sodium cyanide slowly and in small balanced quantities under strongly alkaline conditions to the diamine and heating the mixture with rapid agitation. The compound is compatible with soap and is capable of sequestering metal ions such as calcium ions.

Acrylic type unsaturated acids may be used advantageously in the preparation of compounds included within the scope of this invention. Several methods of employing those unsaturated acids are possible. In general they are first esterified to form a stable ester. Two mols of the ester may then be reacted directly with a diamine, such as ethylene diamine, according to the following equation:

$$NH_2RNH_2 + 2CH_2{=}CH(CH_2)_nCOOR' \rightarrow R'OOCCH_2(CH_2)_{n+1}NRNH(CH_2)_{n+1}CH_2COOR'$$

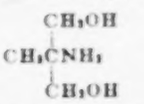
It is noted that the ester group is sufficiently stable so that the hydrogen of the primary amino group acts as a hydrogen donor saturating the double bond. The remaining hydrogen of the amino groups is then reacted according to either of the methods described above. R in the case just described may be $-(CH_2)_n-(NH(CH_2)_n)_m-$ where $n$ is 2 or 3 and $m$ is 0, 1 or 2. A second general method of utilizing an unsaturated ester is hydro-halogenation in the presence of methyl alcohol using dry HCl to form the chloride ester. The halogen ester is then reacted with a polyamine in methyl alcohol in the ratio of two mols of ester to one of the polyamine as in the previous cases. It will be noted that only the primary hydrogens are treated as being in the A positions.

In addition to the compounds described above derivatives using the condensation products from the Diels-Alder reaction may be used. A preferred compound of this series is produced from maleic anhydride and butadiene and is represented by the following formula:

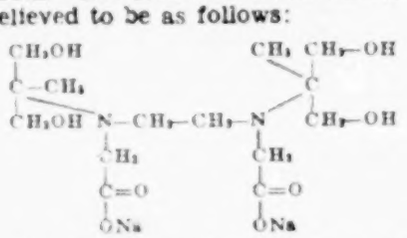


The dicarboxylic acid resulting from the condensation of the butadiene and maleic anhydride is condensed with ethylene diamine in the same manner that the unsaturated acids described above are condensed, that is, the di-acid is esterified and is reacted with hydrochloric acid and the hydrochloride is in turn reacted with ethylene diamine or other suitable diamines, triamines or tetramines. As in the other compounds described, the secondary hydrogens attached to the nitrogen groups are reacted to substitute acetic acid or propionic acid radicals. The product is compatible with soap and is an excellent cleaning agent. If desired, the ester may be saponified to yield the alkali metal salt, and if the acid is preferred, the salt is treated with an acid in the conventional manner.

Still other types of compounds may be produced by reacting 2 amino, 2 methyl, 1-3 propane diol, the formula of which is



with ethylene dichloride. The compound is 2 amino, 2 methyl, 1-3 propane diol is dispersed in methyl alcohol. To this well dispersed, well agitated suspension two mols of ethylene dichloride are added to each ten mols of the amine. The mixture is refluxed for about 1 hour. Sodium carbonate is added from time to time to maintain the pH between 8.0 and 8.5. The reaction is over in approximately 5 hours and requires about two mols of sodium carbonate and 1500 cc. of methyl alcohol. Sodium chloride formed is removed by filtration and the alcohol is removed by distillation. Secondary hydrogen atoms on each of the nitrogen atoms are reacted according to my U. S. Patent 2,407,645 filed June 21, 1943, and are substituted by the radical $CH_2COONa$. The formula of the resulting product is believed to be as follows:



The product as produced is a yellow-white solid. It has been discovered that if the amine diol is reacted according to the chloracetic acid method some of the hydroxyl groups are involved to form ether acids. The compound forms a clear calcium complex and is an excellent detergent. While it increases the foam of soap solutions it, by itself, is not a foaming agent as are the other compounds described above.

A preferred type of compound of this invention is the derivative of ethylene diamine in which a long chain acid such as undecylenic acid is substituted in the A positions. Two compounds using undecylenic acid are of particular interest and the method of making them is described below.

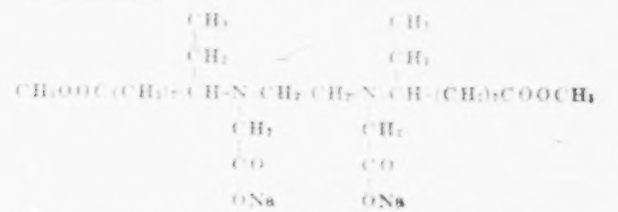
The methyl ester of the acid is first produced. This is in turn reacted with hydrobromic acid. This halogenation is carried out in ligroin in the presence of ar and at a temperature of 0° C. Two mols of the bromide ester react readily with ethylene diamine in the presence of methyl alcohol as a solvent. The reaction is carried out at a pH of 8.0 which is regulated by the addition of dry caustic soda. Sodium bromide is precipi-

tated and may be filtered off. When 70% ethylene diamine is used some foaming will be experienced due to the water present. It has been found that the best procedure is to dehydrate the entire reaction mixture, after the reaction is completed, with benzene. There is no appreciable hydrolysis of the methyl ester during this reaction. After removing the alcohol by distillation, a compound is obtained of a yellowish semi-crystalline mass, soluble in organic solvents and miscible with water to form a milky solution. To react the secondary hydrogen atoms on the nitrogen atoms, the product produced above is redissolved in methyl alcohol and the alcohol solution added to a slurry of chloracetic acid sodium salt in methyl alcohol. Two mols of such salts are used per mol equivalent of the reaction product. There is considerable heat of reaction but if this is permitted to run its course using 200 cc. of methyl alcohol for each mol of chloracetic acid sodium salt, the heat of reaction is 60° C. After two hours sodium bromide begins to precipitate and heat is applied to maintain the reflux temperature of methyl alcohol for about six to eight hours. Sodium carbonate is added from time to time to regulate the pH between 7.5 and 8.5. Provision must be made to allow liberated carbon dioxide to escape. When the reaction is completed the water that is present due to the hydrous diamine is removed before filtering.

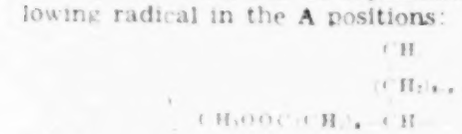
When the undecylenic acid used in the preparation of the foregoing product is of the usual type,

$$CH_3—CH=CH—(CH_2)_7—COOH$$

(9-hendecenoic acid), the product is the dimethyl ester of ethylene diamine N,N'di(sodium acetate) N,N'di(9-ethyl nonylic acid) having the following formula:



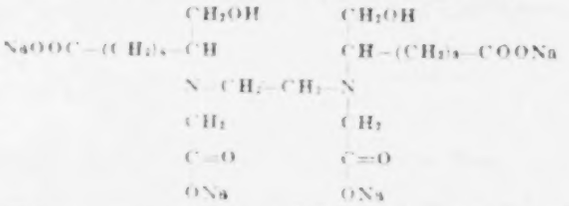
It is obvious that the lengths of the two alkyl chains on either side of the carbon atom of the undecylenic acid ester radical attached to the nitrogen atoms will depend upon the location of the double bond in the undecylenic acid, as well as upon which carbon atom the halogen atom is attached. Hence a more general formula for these compounds, using the methyl ester of any undecylenic acid, will be represented by the following radical in the A positions:



wherein, conformably with the requirements of the compounds of this invention, $x$ is not less than 3, and is, of course, not greater than 8.

The above product is illustrative of these highly effective compounds. They act as cleaners and foam in any type of water. They are not precipitated or salted out by any amount of electrolyte and are powerful wetting agents even in water of 400 parts per million hardness and in sea water. They are compatible with soap and are not precipitated in the form of lime soap nor is the soap salted out. They have a unique dual action in that they will form highly soluble filtrable calcium complexes. They also have the property of dispersing lime salts in an amount far greater than that calculated to form the clear complex. The calcium complex itself is soluble in alcohol. It may be noted that the compounds of this invention are both anionic and cationic in nature. For these reasons, and because these compounds are extremely stable in both alkaline and acid solutions, they have wide usefulness.

A second, and slightly different type of compound from that described above may be prepared from omega undecylenic acid by reacting the acid ester with hypochlorous acid, HOCl. This acid acts much the same way as does hydrochloric acid in its addition to the double bond of undecylenic acid. The addition product is reacted with ethylene diamine and the secondary hydrogen atoms are reacted as described above; upon hydrolysis of the ester groups with alkali a compound of the following formula results;

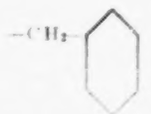


This compound is very similar in nature to the compound described above except that it is more soluble in water. Likewise, the lengths of the two alkyl chains of the undecylenic acid radical will depend upon conditions as set forth above.

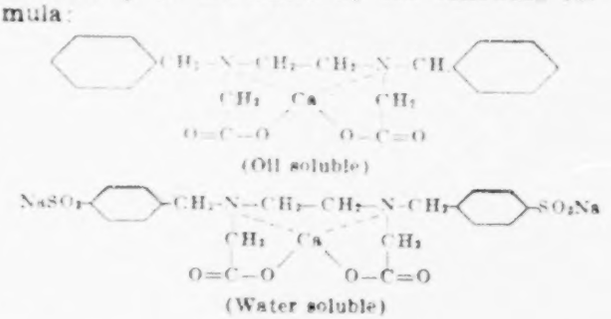
When the hydrochloric acid derivative of undecylenic acid ester is reacted with diethylene triamine, and the secondary hydrogen atoms on the three nitrogen atoms are reacted with chlorpropionic acid sodium salt, there is produced a compound similar to the ethylene diamine derivative wherein the chloracetic acid sodium salt is used. The product is, however, more soluble in water.

The resulting esters formed by the reactions above may be hydrolyzed (as shown in the compound above) or may be retained. Ester groups tend to decrease the solubility of the compound in water.

If the A groups are insoluble or immiscible in water, the calcium complex of the compound is not soluble in water and precipitates. For instance if A is

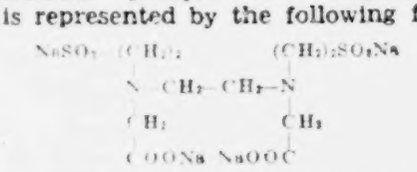


the calcium complex precipitates in water. However, if this benzene nucleus is sulfonated the sodium sulfonate calcium complex is soluble and is useful for the purpose of this invention. If not sulfonated the compound is quite oil soluble, while the sulfonated compound (sodium salt) is quite water soluble. The compounds described above may be illustrated by the following formula:



This compound illustrates the fact that materials particularly suitable for cleaning action and sequestering ability in oil may be secured, by substituting suitable oil soluble groups in the A positions.

The introduction of a (—$SO_3Na$) group into the compounds of this invention may be readily accomplished by reacting two mols of a compound such as taurine, $NH(CH_2)_2SO_3H$, with each one mol of dichloro ethylene, $Cl(CH_2)_2Cl$. The reaction follows the general procedure outlined for that described of reacting 2 amino, 2 methyl, 1-3 propane diol with ethylene dichloride described above. The resulting product is then reacted according to my U. S. Patent 2,407,645 filed June 21, 1943, to substitute the secondary hydrogen atoms on the nitrogen atoms with (—$CH_2COONa$) groups. The completed compound is represented by the following formula:



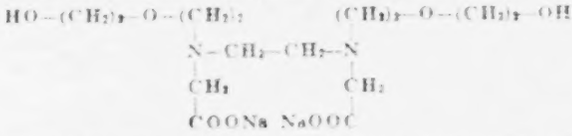
The product may be crystallized from methyl alcohol solutions.

It is excellent as a detergent and for sequestering metal ions, and is completely miscible with soap.

Other types of radicals in the A positions are also advantageous for some cleaning purposes; for example, compounds prepared by reacting an hydroxylated ether with a diamine. The reaction between diethylene glycol and anhydrous ethylene diamine may be carried out merely by boiling the two together. Xylene or a dehydration catalyst may be added but such precaution is not essential. As in the previous examples, the secondary hydrogen atoms on the nitrogen atoms are substituted with —$CH_2COONa$ or

—$CH_2CH_2COONa$

groups, and by the same method. The final compound may be represented by the formula:



This compound is an excellent detergent and sequestering agent and is completely miscible with soap.

A summary of the properties of the compounds herein described will serve to point out obvious uses for them.

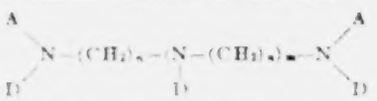
As prepared, the compounds have no deleterious electrolytic properties to soap. Accordingly as they are compatible with soap when incorporated with soap in which electrolytes are present, the electrolytes do not adversely affect the soap. They are of themselves good wetting agents and detergents but do not defat the skin. Being completely stable in both alkali and acid solutions, they may be incorporated in media of either type. The effectiveness of these compounds may be due, in part, to the fact that they are both cationic and anionic in action as well as the fact that they sequester metal ions, such as calcium ions, thus preventing the precipitation of calcium soaps. It can be seen from the above description that the compounds may be varied to make them soluble in oils, organic solvents, or water. The foaming power of the compound may be adjusted, as the longer the aliphatic chain in the A positions, the greater the foaming power, while the heterocylic compound shown has very little foaming action (although it is a good detergent). More than six carbon atoms in the hydrocarbon chain insures the compound of having good wetting action, particularly if carboxyl groups or hydroxyl groups are present.

Although any desired amount of these compounds may be added to a water-soluble soap to form a washing compound, it has been found that from 1% to 5% by weight prevents the precipitation of calcium or magnesium soaps in any commonly used water, and yields a good detergent soap.

This application is a continuation-in-part of my copending application Serial No. 657,893, filed March 28, 1946, which issued as Patent No. 2,524,218 on October 3, 1950.

I claim:

1. A compound having the formula



wherein $n$ is an integer selected from the group consisting of 2 and 3; $m$ has a value equal to one of the group consisting of zero, the numeral 1, and any multiple thereof; D is a member of the group consisting of —$CH_2COOH$,

—$CH_2 \cdot CH_2 \cdot COOH$

and their alkali metal salts; A is an alkyl group containing at least seven carbon atoms and having not more than two hydrogen atoms thereof replaced by a member of the group consisting of —COOR, —COOM, —$SO_3H$, —$SO_2M$, —OR, and —OR′OR wherein R is one of the group consisting of alkyl and hydrogen, M is an alkali metal, and R′ is an alkylene group, each said replacing groups being spaced from the N atoms of the compound by at least two carbon atoms of the hydrocarbon group containing the same.

2. The compound of N,N di-octyl ethylene diamine N,N′ di-acetic acid.

3. The compound ethylene diamine N,N′ di-acetic acid N,N′ di (9-ethyl nonylic acid).

FREDERICK C. BERSWORTH.

REFERENCES CITED

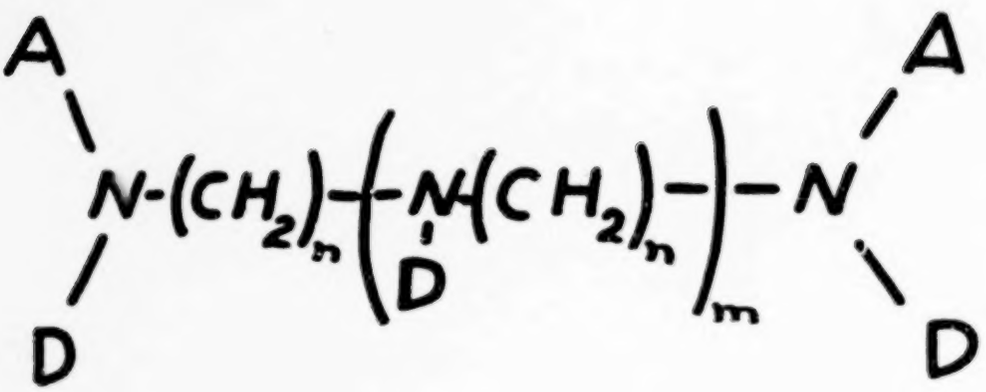
The following references are of record in the file of this patent:

UNITED STATES PATENTS

| Number | Name | Date |
|---|---|---|
| 2,130,505 | Munz | Sept. 20, 1938 |
| 2,240,957 | Munz | May 6, 1941 |
| 2,293,034 | Moore | Aug. 18, 1942 |

Plaintiffs' Exhibit No. 3 C. A. 1637-54

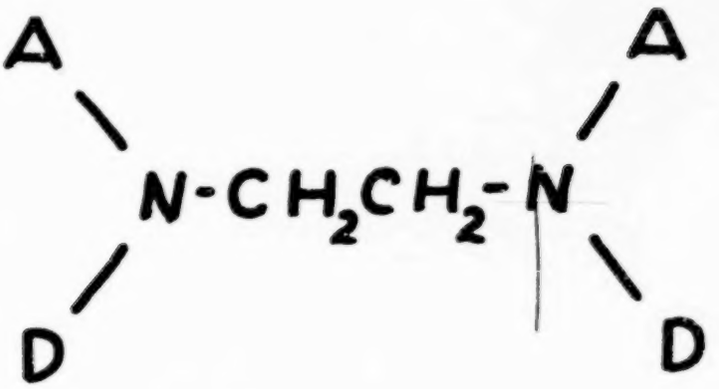
CASE-A
U.S. PAT. 2,524,218



Plaintiffs' Exhibit No. 4 C. A. 1637–54

**CASE-A**

U S PAT. 2,524,218

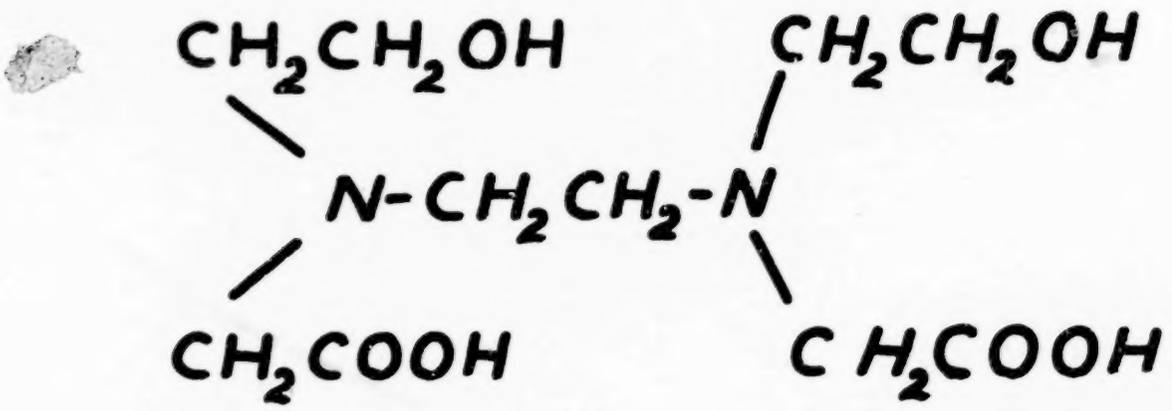


Plaintiffs' Exhibit No. 5

C. A. 1637–54

CASE - A

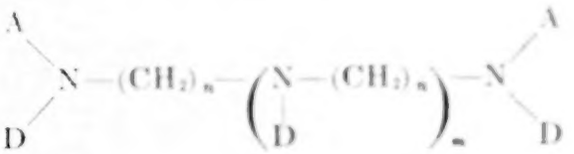
U.S. PAT. 2,524,218



*Excerpts From Plaintiffs' Exhibit No. 6*

I CLAIM:

1. A compound having the formula



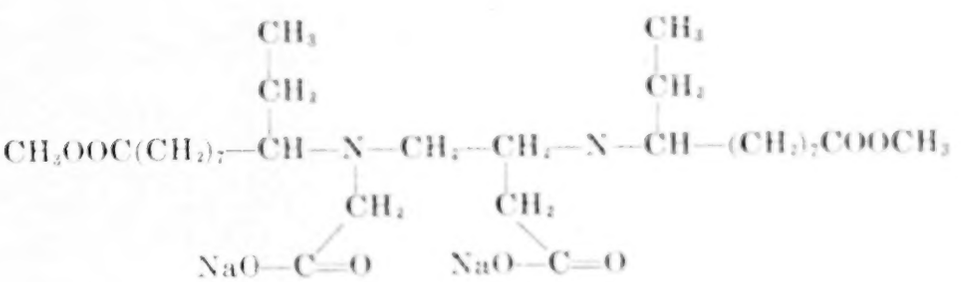
wherein n is either 2 or 3 and m is either zero or a positive integer and wherein D is a member of the group consisting of $-CH_2COOH$, $-CH_2CH_2COOH$, and their alkali metal salts, and wherein A is a hydrocarbon group containing at least two carbon atoms and having not more than two hydrogen atoms replaced by a member of the group consisting of $-(CH_2)_2COOR$, $-(CH_2)_2COOM$, $-SO_3H$, $-SO_3M$, $-OR$, and $-OR'OR$ wherein R is alkyl or hydrogen, M is an alkali metal, and R′ is alkylene, said groups being spaced at least two carbon atoms from the nitrogen atoms.

2. A compound according to Claim 1 wherein D is the radical $-CH_2COOR$, R being hydrogen or alkali metal.

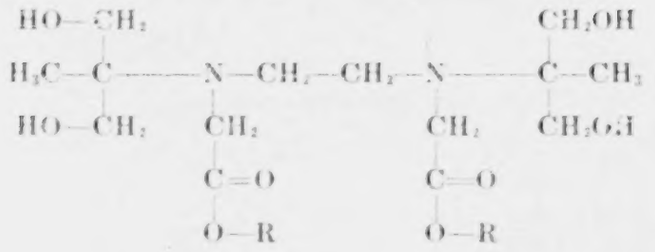
3. A compound according to Claim 1 wherein the hydrocarbon groups are alkyl groups.

4. A compound according to Claim 1 wherein the hydrocarbon groups are cyclic groups.

5. A compound having the formula

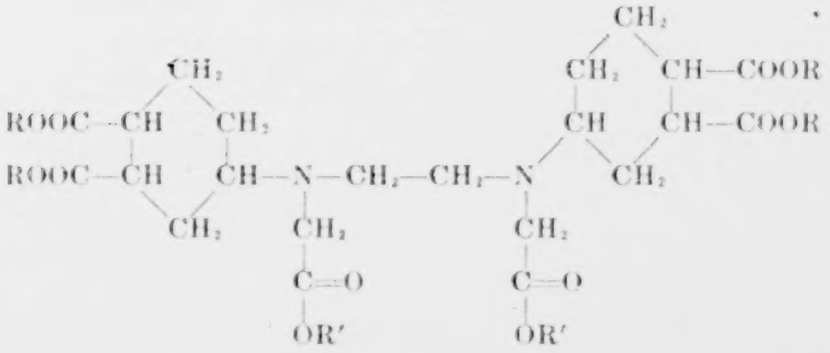


6. A compound having the formula



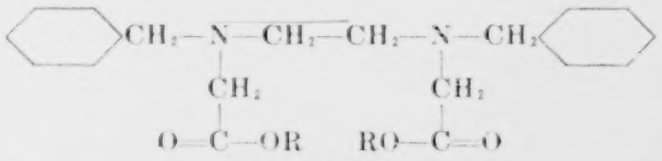
wherein R is a hydrogen or an alkali metal.

7. A compound having the formula



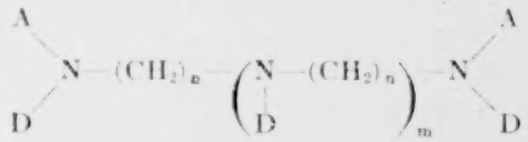
wherein R is hydrogen or an alkyl group and R′ is hydrogen or an alkali metal.

8. A compound having the formula



wherein R is hydrogen or an alkali metal.

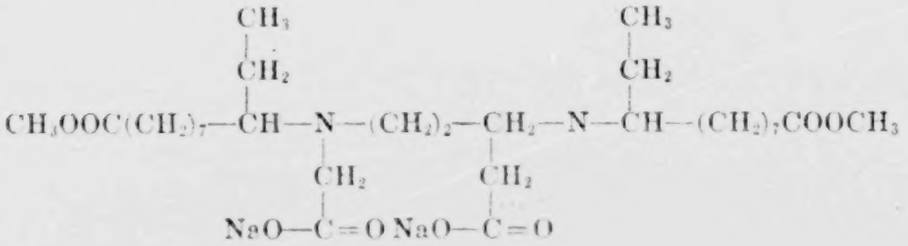
9. As a washing composition a mixture comprising as its essential active ingredients a water soluble soap and a compound having the general formula



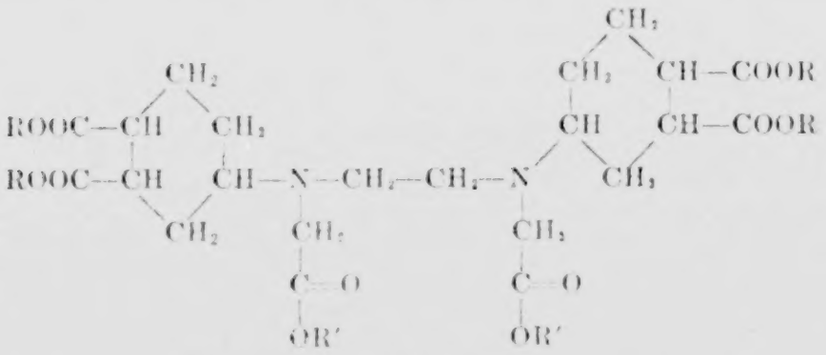
wherein n is a positive whole number from 2 to 3 and m is zero or a positive integer and wherein D is

a member of the group consisting of $-CH_2COOH$, $-CH_2CH_2COOH$, and their alkali metal salts, and wherein A is a hydrocarbon group containing at least two carbon atoms and having not more than two hydrogen atoms replaced by a radical of the group consisting of $-(CH_2)_2COOR$, $-(CH_2)_2COOM$, $-SO_3H$, $-SO_3M$, $-OR$, and $-OR'OR$ wherein R is alkyl or hydrogen, M is an alkali metal, and R′ is alkylene, said radical being spaced at least two carbon atoms from the nitrogen atoms.

10. As a washing composition, a mixture of a water soluble soap and a compound having the formula

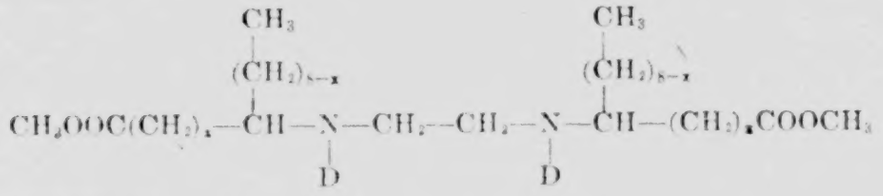


11. As a washing composition, a mixture of a water soluble soap and a compound having the formula



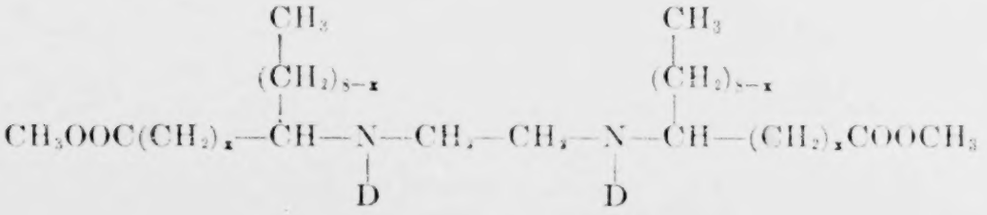
wherein R is hydrogen or an alkyl group and R′ is hydrogen or an alkali metal.

12. A compound having the formula



wherein x is a positive integer between 2 and 8, inclusive, and D is a member of the group consisting of $-CH_2COOH$, $-CH_2CH_2COOH$, and their alkali metal salts.

13. A washing composition having as its essential active ingredients a water-soluble soap and a compound having the formula



wherein x is a positive integer from 2 to 8, inclusive, and D is a member of the group consisting of $-CH_2COOH$ $-CH_2CH_2COOH$, and their alkali metal salts.

Signed at Cambridge, Massachusetts, this 27th day of March 1946.

FREDERICK C. BERSWORTH.

*Oath*

COMMONWEALTH OF MASSACHUSETTS,

*County of Middlesex,*

Frederick C. Bersworth, the above named petitioner, being duly sworn, deposes and says that he is a citizen of the United States, and a resident of Verona in the County of Essex and State of New Jersey; that he verily believes himself to be the original, first, and sole inventor of the improvements in Alkylene polyamine derivatives described and claimed in the annexed specification; that he does not know and does not believe that the same was ever known or used before his invention or discovery thereof, or patented or described in any printed publication in any country before his invention or discovery thereof, or more than one year prior to this application, or in public

use or on sale in the United States for more than one year prior to this application; that said invention has not been patented in any country foreign to the United States on an application filed by him or his legal representatives or assigns more than twelve months prior to this application; and that no application for patent on said improvements has been filed by him or his representatives or assigns in any country foreign to the United States.

FREDERICK C. BERSWORTH.

Sworn to and subscribed before me this 27th day of March 1946.

[SEAL] ALLAN L. SPURR,
*Notary Public.*

My commission expires Nov. 14, 1952.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington 25, D. C.

Div. 64, Room 7600 Paper No. 3

Address only: The Commissioner of Patents, and not any official by name.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the examiner in charge of this application. Casper W. Ooms, Commissioner of Patents.

Applicant: Frederick C. Bersworth. Ser. No. 657,893. Filed Mar. 28, 1946. For Alkylene Polyamine Derivatives.

Frank N. Houghton,
35 East St., Elmwood, Mass.

References:
Mumz—2,240,957, May 6, 1941, 252–117.

Bersworth—2,387,976, Oct. 30, 1945, 260–539.

The claims herein are drawn to two separate and distinct alleged inventions as follows:

(I) claims 1–8 and 12 are drawn to a chemical compound which is classifiable in class 260 as shown in the patent to Bersworth cited above.

(II) claims 9–11 and 13 are drawn to a detergent composition which is classifiable in class 252 as shown by Munz cited above.

The two groups are shown to separate and distinct alleged inventions which are separately classified and searched and each of which has acquired a separate status in the art as shown by the references cited. Moreover the compounds of group (I) are capable of many uses such as detergents, wetting agents, and sequestering of calcium ions. The claims of group (II) are drawn to one of such uses.

Division is accordingly required between (I) claims 1–8 and 12 and (II) claims 9–11 and 13.

The references cited above are a result of a cursory search of the art. Action on the merits of the claims will be given upon compliance with the requirement of division.

P. M. NASH,
*Examiner.*

JG: as.

In the United States Patent Office

Division 64—Room 7600

ELMWOOD, MASSACHUSETTS, *December 20, 1946.*

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 657,893. Filed March 28, 1946. For: Alkylene Polyamine Derivatives.

Hon. COMMISSIONER OF PATENTS,
*Washington, D. C.*

SIR: Responsive to the Office Act of July 8, 1946, requiring division, the following remarks are submitted:

REMARKS

Reconsideration of the requirement for division between Claims 1–8 and 12 drawn to a chemical compound and Claims 9–11 and 13 drawn to a detergent composition, is respectfully asked.

Requirement for division is apparently based upon Office Classification, and although such a basis is recognized as being presumptive of different inventions, it is by no means controlling. *United States ex rel Steinmetz* v. *Allen*, 192 U. S. 543.

A survey of the claims will show that Claims 9–11 and 13 (Group II) directed to a washing compound are, in effect, the compounds of Group I diluted. For instance, Claim 9 is directed to a washing composition having the compound of Claim 1, and similarly Claim 10 parallels Claim 5, Claim 11 parallels Claim 7, and Claim 13 parallels Claim 12. Under such conditions it might be later found that claims in Group II would be unpatentable over those of Group I. In *Ex parte Nichols*, 23 USPQ 94, it was said that, where by requirement for division the validity of granted patents may be jeopardized, division should not be insisted upon.

It has been said that requirement for division should not be insisted upon if one set of claims would infringe a second set,–Ex parte Hunter 25 U. S. P. Q. 24. The manufacture of washing compounds of group II would infringe the claims of group I.

It is respectively submitted in view of the remarks submitted above, the requirement for division should be withdrawn.

In the event that the requirement for division should be made final, applicant elects for prosecution on the merits those claims falling within group II, namely, claims 9, 10, 11 and 13.

Respectfully,

FREDERICK C. BERSWORTH,
By FRANK N. HOUGHTON,
*His Agent.*

MARCH 20, 1947.

IN RE APPLICATION OF F. C. BERSWORTH :

Serial No. 657,893. Filed: March 28, 1946. For: Alkylene Polyamine Derivatives.

Examiner, Division 64:

Division between:

(I) Claims 1–8, 12

(II) Claims 9–11, 13 is approved.

The two groups are directed to distinct inventions, separately classified and examined.

The compound of group I is stated to be of wide utility, and is not restricted to use in a washing composition also containing a water soluble soap as defined in the claims of group II.

The election of group II is noted.

Respectfully,

G. A. GORECKI,
*Acting Examiner of Classification.*

GG: jam.

DEPARTMENT OF COMMERCE

UNITED STATES PATENT OFFICE

Washington, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Div. 64, Room 7600

Paper No. 5

Address only: The Commissioner of Patents, and not any official by name.

Please find below a communication from the examiner in charge of this application. Casper W. Ooms, Commissioner of Patents.

Applicant: Frederick C. Bersworth. Ser. No. 657,893. Filed, Mar. 28, 1946. For Alkylene Polyamine Derivatives.

FRANK N. HOUGHTON,

*35 East St., Elmwood, Mass.*

Responsive to communication of December 23, 1946.

Additional references:

Bersworth—2,412,945, December 24, 1946, 252–117.

This application was submitted to the Examiner of Classification whose decision is as follows:

Division between:

(I) Claims 1–8, 12

(II) Claims 9–11, 13 is approved.

The two groups are directed to distinct inventions, separately classified and examined.

The compound of group I is stated to be of wide utility, and is not restricted to use in a washing composition also containing a water soluble soap as defined in the claims of group II.

The election of group II is noted.

The requirement for division is accordingly repeated and now made Final.

It is noted that applicant has elected to prosecute the claims of group (II); namely, 9, 10, 11, and 13. Claims 1–8 and 12 are accordingly rejected on the ground of misjoinder of invention.

Claim 9 is rejected as indefinite and alternative in the compound having the general formula recited therein. The values of the various letters are alternative.

Claim 9 is also rejected as indefinite and too broad in the compound recited therein. Such values as "a positive integer", "alkyl" and "alkylene" are indefinite in scope.

Claims 9 and 13 are rejected on the ground of insufficient disclosure. No specific example of a water-soluble soap is found in the disclosure.

Claim 13 is also rejected as indefinite and too broad as to the ingredients of the composition intended to be covered by the claim.

Claims 9 and 13 are also rejected as lacking invention over the patent to Bersworth cited above which shows a detergent composition consisting of a mixture of an alkali metal fatty acid soap and a compound of the type recited in the claims.

Claims 10 and 11 are drawn to two different species of the alleged generic invention disclosed. The generic claims having been rejected, election of a single species for further prosecution is required.

Applicant's response to be complete must include an amendatory action or traverse relating to the rejected generic claims and an election of species, con-

formably to the Examiner's requirement. Order No. 3527, Oct. 24, 1940; 520 O. G. 259.

P. M. NASH,
*Examiner.*

JG:nks.

In the United States Patent Office

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 657,893. Filed: March 28, 1946. For: Alkylene Polyamine Derivatives.

Division 64—Room 7600

ELMWOOD, MASSACHUSETTS, *May 19, 1947.*

Hon. COMMISSIONER OF PATENTS,
*Washington, D. C.*

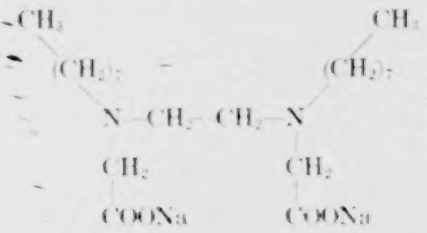
SIR: Responsive to the Office Action of April 7, 1947, please amend as follows:

Cancel claims 1–8 and 12.

Add the following claims:

—14. A washing composition according to claim 9 wherein A is a hydrocarbon group containing at least 7 carbon atoms and having not more than two hydrogen atoms replaced by a radical of the group consisting of –COOR, –COOM, $-SO_3H$, $-SO_3M$, –OR, and –OR′OR wherein R is a member of the group consisting of alkyl and hydrogen, M is an alkali metal, and R′ is an alkylene group, said radical being spaced at least two carbon atoms from the nitrogen atoms.

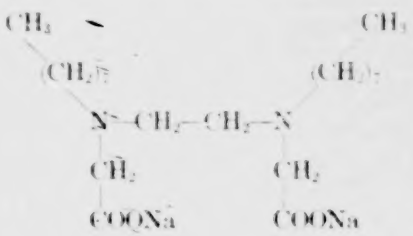
—15. A washing composition according to claim 9 wherein the compound has the formula



In the specification change the title to read "Washing Compositions".

Page 2 line 15; page 3, line 8; page 5, line 4; and page 7, line 4; after "1943" insert —, now U. S. Patent 2,407,645—

Page 4 cancel the formula and insert—



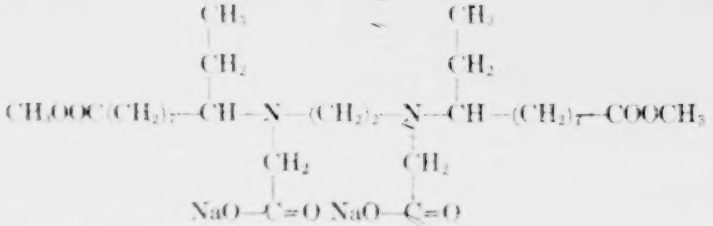
Page 10 line 24, after "491,669" insert —, now U. S. Patent 2,407,645—

Claim 9, in the first line after the formula cancel "either 2 or 3 and m is either", and insert —a positive whole number from 2 to 3 and m is—

in the fifth line after the formula cancel "member" and insert —radical—

in the eighth line after the formula cancel "groups" and insert —radical—

Claim 10, cancel the formula and insert the following formula:



Claim 13, in the first line after the formula cancel "between 2 and 8" and insert —from 2 to 8—

## Remarks

Claims 1–8 and 12 have been cancelled leaving only those claims i. e., 9, 10, 11, and 13 directed toward the elected invention.

The formula on page 4 has been corrected to rectify a typographical error.

Claim 10 has been amended to correct a typographical error so that it now conforms to formula given on page 8 as was originally intended.

The new claims 14 and 15 have been added to cover more adequately the invention made. Support in the specification may be found on pages 3 and 4.

Claim 9 was rejected as alternative and indefinite in the compound having the general formula therein recited. The claim has been amended to more clearly define the term "n". The claim is now believed to be free of the difficulty of being either unduly indefinite or unduly alternative. The terms "a positive integer", "alkyl" and "alkylene" are well known and commonly used terms having a definite meaning to those familiar with organic chemistry. The attention of the Examiner is called to U. S. Patent 2,396,938 issued to Frederick C. Bersworth, the inventor of the present invention. The formulas in several of the claims of that patent and the description of the terms closely parallel those here submitted. If the rules of the Office have been altered in the past year, or if the claims of that patent are clearly not in proper form, the applicant would appreciate a statement by the Examiner to that effect. It is believed that consistency in the Patent Office is necessary for the proper prosecution and issuance of patents unless special circum-

stances require a change in ruling. In such case the reasons requiring such change should be clearly set forth by the Examiner. A general description of the compounds of the present invention is difficult, and applicant would welcome any suggestions as to improvement in form.

Claims 9 and 13 are rejected as having no specific example of a water soluble soap in the disclosure. Soaps are broadly divided into two types, i. e., water soluble and water insoluble, and examples of soaps falling in each of these classifications are well known. The specification clearly indicates that the mixture contemplated is the water soluble variety of soap. Perhaps the principal use of the compositions of this invention is when mixed with ordinary soap; the composition being present to prevent the formation of hard-water-insoluble soaps. Such use is clearly indicated by the examples in the specification. Further, the last paragraph on page 12 states specifically that "these compounds may be added to a water-soluble soap." Where the members of the generic term are so well known, and where the particular members are not in any way critical, a specific example would appear to be superfluous.

The Examiner has rejected claim 13 as indefinite and too broad. As amended applicant believes that there is nothing indefinite about the composition. It is a *washing composition* having as active ingredients *a water soluble soap* and a compound having *a given chemical formula*. The basis for this claim is given on pages 8 and 8a and the reason for writing the undecylenic acid reaction product in such a fashion is because of the location of the double bond which is different for different undecylenic acids. If the Examiner still believes that the claim is either indefi-

nite or too broad, or both, a fuller statement of his objections would be useful.

Claims 9 and 13 are rejected on U. S. Patent 2,412,946 to Bersworth. The compounds of that patent result from the reaction of an aliphatic polyhydroxy alcohol and an alkali metal salt of a lower alkyl amino polyacetic acid. The compounds of the present invention are not polyacetic acid derivatives reacted with polyhydroxy alcohols. As explained in the specification (see p. 3 et seq.), the compounds are directed toward longer chain hydrocarbon radicals in the "A" positions of the formula. Such compounds are more soap-like and are more compatible with soap than those containing acetic acid radicals. A further difference is that the present invention does not contemplate reacting the alkali metal salt of an amino acid with a polyalcohol. It is believed that the claims clearly distinguish over U. S. 2,412,945.

Applicant elects the species represented by claim 10. It is requested, however, that the claim 11 be retained in the pending determination of the generic claims.

All the remaining claims are now believed to be in condition for allowance. Allowance is accordingly solicited.

Respectfully,

FRANK N. HOUGHTON,
*Agent for Applicant.*

DEPARTMENT OF COMMERCE

UNITED STATES PATENT OFFICE

Washington

Paper No. 7

Address only: The Commissioner of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the EXAMINER in charge of this application. Lawrence C. Kingsland, Commissioner of Patents.

Division: 64. Applicant: Frederick C. Bersworth. Ser. No. 657,893. Filed Mar. 28, 1946. For Washing Compositions.

FRANK N. HOUGHTON,
*35 East St., Elmwood, Mass.*

Responsive to amendment of May 20, 1947.

Additional references:

Bersworth—2,407,645, Sept. 17, 1946 (Filed 6–21–43), 260–534.

Munz—2,240,957, May 6, 1941, 252–117.

The claims herein are now 9–11 and 13–15.

It is noted that applicant has elected the species represented by claim 10. Claim 11 to non-elected species is accordingly rejected as not readable on the elected species.

Claim 9 is rejected as indefinite and too broad as to the compound having the general formula recited in the claim. The values of "M" and "R" are alternative. The term "alkyl" is indefinite as to scope.

Claims 9, 10, and 13–15 are rejected on the ground of insufficient disclosure in "a water-soluble soap." No specific example thereof is found in the disclosure.

Claims 9 and 13–15 are also rejected as indefinite and too broad as to the ingredients of the composition other than those specified in the claims. The claims merely set forth the "active ingredients."

Claims 9, 10, and 13–15 are also rejected as too broad in "a water-soluble soap." Soaps differ materially in composition and properties.

Claims 9, 10, and 13-15 are also further rejected as not properly defining the alleged invention by failing to set forth the relative proportions in which the ingredients are present

Claims 9, 10, and 13–15 are also rejected as lacking invention over the patent to Bersworth of record or Bersworth or Munz cited above. The use of a compound of the type claimed together with soap would not involve invention over either of the patents which discloses a mixture of a fatty acid soap and an alkylene polyamine derivative.

All the claims are rejected.

P. M. NASH,
*Examiner.*

JG:nks.

In the United States Patent Office

Applicant: Frederick C. Bersworth. Invention: Washing Compositions. Division 64, Ser. No. 657,893. Filed: Mar. 28, 1946.

NEW POWER OF ATTORNEY

Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

Sir: In the above identified application I hereby revoke all previous powers of attorney and appoint Malcolm N. Rich (Reg. No. 13,142), of 11 Park Place, New York, 7, New York, as my attorney with full power to prosecute said application, to make alterations and amendments therein, to receive the patent, and to transact all business in the Patent Office connected therewith.

Signed at Framingham, Mass., this 17th day of May, 1948.

FREDERICK C. BERSWORTH,
Frederick C. Bersworth,
*Applicant.*

MNR: W.

DEPARTMENT OF COMMERCE

UNITED STATES PATENT OFFICE

Washington

JUNE 3, 1948.

MALCOLM N. RICH,
*11 Park Place, New York 7, N. Y.*

Applicant: Frederick C. Bersworth. Serial No. 657,893. Filed March 28, 1946. For Washing Compositions. Div. 64.

In this case your power of attorney has been accepted.

Very truly yours,

LAWRENCE C. KINGSLAND,
*Commissioner of Patents.*

Revoking power of attorney to Frank N. Houghton, 35 East Street, Elmwood, Mass.

In the United States Patent Office

Division 64. Ser. No. 657,893. Filed Mar. 28, 1946.
Applicant: Frederick C. Bersworth.
Invention: Washing Compositions.

AMENDMENT (3)

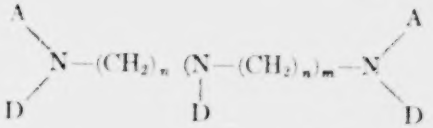
Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: In response to the Official Action of January 26, 1948, please amend the above application as follows:

CLAIMS

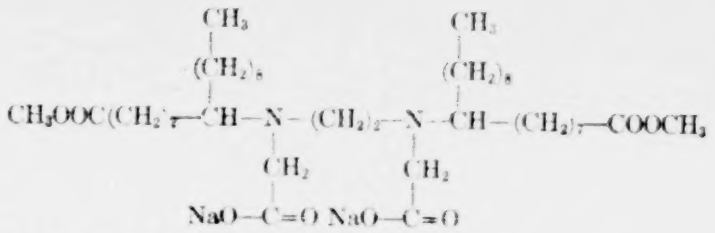
Erase all claims of record and substitute therefor the following claims:

1. A detergent composition consisting of a water soluble alkali metal-fatty acid soap and an alkylene polyamine derivative, the amount of said alkylene polyamine derivative in the said mixture being from 1% to 5% by weight of the mixture, said alkylene polyamine derivative being one conforming to the formula:



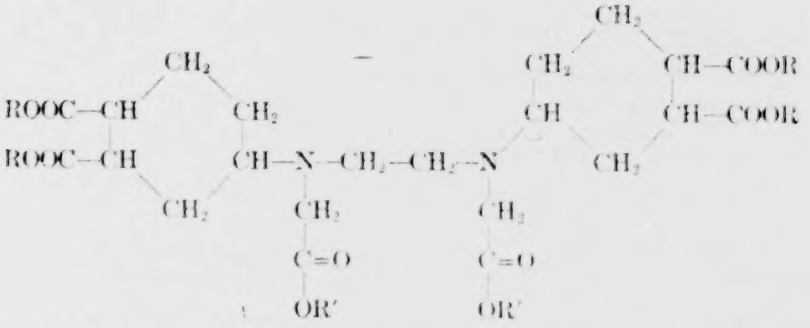
wherein $n$ is an integer selected from the group consisting of 2 and 3; $m$ has a value equal to one of the group consisting of zero, the numeral one (1), and any multiple of the numeral one (1); $C_nA$ is a hydrocarbon group containing at least two carbon atoms and having not more than two hydrogen atoms replaced by a member of the group consisting of $-(CH_2)_2COOR$, $-(CH_2)_2COOM$, $-SO_3H$, $-SO_3M$, $-OR$, and $OR'OR$ wherein R is one of the group consisting of alkyl and hydrogen, M is an alkali metal, and R′ is an alkylene group, said replacing groups being spaced from the N atoms of the compound by at least two carbon atoms of the hydrocarbon group containing the same.

2. The detergent composition of claim 1, wherein said alkylene polyamine derivative comprises the compound:



405368—56——12

3. The detergent composition of claim 1, wherein said alkylene polyamine derivative comprises the compound:



wherein R is one of the group consisting of hydrogen and alkyl and R′ is one of the group consisting of hydrogen and alkali metal.

## Remarks

The rejected claims have all been erased and the subjects matter thereof rewritten and reinserted as new claims 16–19 inclusive. It is believed that the new claims are free of the form defects pointed out by the Examiner as to the erased claims.

In the new claims, claim 16 is generic and claims 17 to 19, inclusive, are species claims depending from generic claim 16.

Generic claim 16 defines the present invention as a detergent composition comprised of a *mixture* of two ingredients. One ingredient is identified as a water soluble soap compound and the other ingredient is identified as an alkylene polyamine derivative conforming to the generic formula given. In conformity with the disclosure on page 12 of the specifications, the amount of the alkylene polyamine derivative present in the mixture has been specified in the claims as being from 1% to 5% by weight of the mixture.

In claim 16, the formal objections to the identification of the values for n, m, A and D heretofore made

by the Examiner have all been met, it is believed, by the wording employed.

The Examiner objects to the term "water soluble soap" as employed in the erased claims on the ground of insufficient disclosure. Attention is directed to the last paragraph on page 12 wherein a water-soluble soap in admixture with the alkylene polyamine derivative compound to form a washing compound, is specified. The Examiner's rejection on this ground, accordingly, is in error.

The Examiner objects (last paragraph on page 1 of the action) to the erased claims being indefinite and too broad. It is presumed by this that the Examiner means that the term "comprising" as used in the erased claims is objected to. It would be helpful if the Examiner would be more specific in this type of rejection, so that the applicant would not have to "guess" at the Examiner's meaning. As worded the new claims are believed free of this objection.

The objection to the term "water-soluble soap" (paragraph 1, page 2 of action) is believed unwarranted. The specification is believed quite clear that the alkylene polyamine derivatives of the present invention are utilizable in association with all of the different types of water soluble soaps. (See last paragraph page 11 of specification.)

Applicant, by stating that the compounds of his invention are "completely stable in both alkali and acid solutions" refutes the assumption by the Examiner that the term "water-soluble soap" is too broad and indefinite. The compounds of the present invention can be used with any kind of water-soluble soap (acid or alkali).

As to patentability, the detergent composition now defined by the new claims 16–19, inclusive, is believed clearly patentable over Bersworth 2,412,945, or over

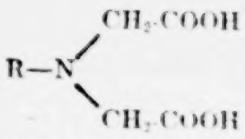
Bersworth 2,407,645, or over Munz 2,240,957 for the primary reason that the *combination* forming the mixture is new and useful and represents an advance in the art of detergents.

As pointed out in the previous amendment the compounds disclosed by Bersworth 2,412,945 and *recited by* the *claims* of this patent are *dissimilar* to the compounds *claimed* by the present invention, and are limited to use in association with alkali metal fatty acid soap compounds. As this patent was *copending* with the present application, it cannot be used by the Examiner for purposes of "anticipation". The rule is, in the case of copending applications, that *different* inventions must be *claimed*.

The instant application is claiming a mixture which clearly is different in composition from that of the patent.

The same remarks apply to Bersworth 2,407,645. The present application was *copending* with this patent. This patent merely discloses and *claims* a process of preparing aliphatic polycarboxylic amino acids which may be employed in the manufacture of the compounds of the present invention. As the *claims* of this patent are *limited* to such process, no question of double patenting arises. The reference is not available to the Examiner for purposes of *anticipation*.

The rejection on Munz 2,240,957 is believed technically unsound. Munz discloses and claims a mixture consisting of a water soluble soap and an *alkali metal salt* of a carboxylic acid of the general formula




wherein R consists of one of three (3) noted radicles. (See claim 6.) None of the radicles specified by this

claim 6 of Munz (or in claim 7 either) meet the definition of the compound of the present invention as now defined in new claim 16.

No *disclosure* in Munz appears whereby one skilled in the art can reasonably be presumed to be taught either how to make the compounds of the present invention or of the utility of the said new compounds in association with water-soluble soap compounds.

Moreover, the new compounds of the present invention react differently from the compounds of Munz as disclosed by applicant on page 11 (last paragraph).

Accordingly, the detergent composition of the present invention is a new *combination* of soap and associated ingredients, functioning in a different manner to produce a desired result.

It is therefore submitted that Munz cannot be held an anticipation of the present invention. As no combination of Munz and the Bersworth patents are available to the Examiner for purposes of anticipation (under the copending application doctrine) the new claims are believed allowable to the applicant for the reasons above stated.

A prompt and favorable action on the new claims is respectfully solicited.

Respectfully submitted.

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His attorney.*

JULY 1, 1948.
MNR: W.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Paper No. 11

Address only: The Commissioner of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the examiner in charge of this application. Lawrence C. Kingsland, Commissioner of Patents.

Division: 64. Applicant: Frederick C. Bersworth. Ser. No. 657,893. Filed March 28, 1946. For Washing Compositions.

MALCOLM N. RICH,
*11 Park Place, New York 7, N. Y.*

Responsive to amendment filed July 2, 1948.

The claims herein are now 16–19.

Claims 18 and 19 to non-elected species are rejected as not readable on the elected species. The species of claim 17 was elected.

Claims 16 and 17 are rejected on the ground of insufficient disclosure. No specific example of a water-soluble soap is given in the original disclosure. The last paragraph on page 12, referred to by applicant merely refers to a water-soluble soap. No examples are disclosed.

Claims 16 and 17 are rejected as indefinite in "comprised of a mixture consisting of". The words "comprised of a mixture" should be cancelled to overcome this ground of rejection.

Claim 16 is also rejected as indefinite as to the alkylene polyamine derivative recited therein. The value of D is not given.

Claims 16 and 17 are also rejected as lacking invention over Munz of record. The use of a compound of the type claimed together with soap would not involve invention over the reference which shows a mixture of a fatty acid soap and an alyklene polyamine derivative.

All the claims are rejected.

This rejection is made final.

No further amendments that do not place this application in condition for allowance or better form for appeal will be entered unless accompanied by a proper showing of good and sufficient reasons why they were not earlier presented. Rule 68 and Commissioner's Notice of April 26, 1948.

P. M. NASH,
*Examiner.*

JG/mk.

In the United States Patent Office

Applicant: Frederick C. Bersworth. Invention: Washing Compositions. Division #64. Serial No. 657,893. Filed: March 28, 1946.

AMENDMENT (4)

FOR PURPOSES OF ALLOWANCE OR APPEAL

HON. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: Please amend the above application as follows:

CLAIMS

Claim 16, lines 1 and 2, erase "comprised of a mixture"; line 3, before "soap" insert—alkali metal-fatty acid —; line 13, after "(1):" insert — D is a member of the group consisting of $-CH_2 \cdot COOH$, $CH_2 \cdot CH_2 \cdot COOH$ and their alkali metal salts;

Claim 17, in the formula, in the carbon chain extending upwardly from the CH group attached to each N, enclose the $CH_2$ group in parentheses and add the subnumeral 8 to make the formula read:

Erase claim 19.

## Remarks

The above amendments to the Finally Rejected claims are made for the purpose of allowance or appeal. The amendments, to each of the claims 16 to 18 inclusive are believed to overcome the Examiner's objection to indefiniteness and on insufficient disclosure as well as incompleteness. Claim 19 has been erased as being a species not included with the generic claim 16.

Reconsideration of the final rejection is respectfully requested in view of the following remarks:

The Examiner relies on Munz holding "no invention" in the use of an alkylene polyamine *derivative* in association with fatty acid soap.

Attention is directed to the fact that the genus and species compounds defined by the rejected claims *are not derivatives* of any of the alkylene polyamines disclosed by Munz.

The alkylene polyamine derivatives of the present invention are new, *per se*. Munz discloses the *acetic acid derivatives* of amines and diamines. The compounds of the instant invention are derivatives of *poly-amines* wherein on *each amino nitrogen only one* (*1*) of the amino hydrogens have been displaced by $CH_2 \cdot COOH$ or $CH_3 \cdot CH_2 \cdot COOH$ groups.

The other amino hydrogen on each amino nitrogen has been displaced by a hydrocarbon group of the type defined in the claims for "A".

In view thereof, it is believed that the Examiner is in error in his holding of non-invention over Munz. As the compounds are new, per se, the *combination* of the same with fatty acid soap compounds necessarily also is new.

It is believed that novelty and patentability in the instant invention falls under the general theorem of patent law that where one of the elements in a combination is new, per se, the combination is necessarily new unless the new element is a *substantial equivalent* for that displaced from the combination.

As to this point the attention of the Examiner is directed to the applicant's disclosure (page 2) to the effect that the present compounds, *contra to those of Munz*, are compatible with fatty acid soap compounds and are, per se, *detergents*.

As *detergents*, per se, the compounds of the present invention in admixture with fatty acid soap compounds from a *different* combination, namely, a *detergent combination or composition*.

In this respect, the composition of the instant invention is an *improvement* of Munz. In such an improvement the obviousness of forming the combination *after* the new compounds of the present invention have been discovered and their properties determined is *not* the test for novelty.

The novelty of the combination rests on the novelty, per se, of the new compounds. It is urged that patentability rests on the old and well recognized rule of patentability that where one of the elements in a combination is new, per se, and not a substantial equivalent for the element displaced in an old combination, the new combination is patentably new, irre-

spective of the obviousness of combination as the patenability of the same rests on the novelty of the new element.

With reference to limitation of the claims to a composition consisting of alkali metal fatty acid soap compounds and the alkylene polyamine derivatives, justification thereof is believed found in the specification, page 8a, lines 14 to 16, page 11, last paragraph, and the last paragraph on page 12. In each of these parts of the specification while the term used is "soaps" it is believed obvious that this term is meant to identify the alkali metal fatty acid soap compounds commonly known in the art under the term "soap" as such compounds are the ones which form *insoluble* calcium or magnesium soaps or soap compounds, i.e., insoluble Ca or Mg salts of the fatty acids.

In this connection it is pointed out that the Munz patent (page 2, column 1, lines 72–75) identifies the term "the soaps" as being the "salts of the higher fatty acids" thus confirming applicant's intended meaning of the term "soap" as it is used in his specification.

In view of the fact that Munz failed to recognize that in his combination the alkali metal salts of the amino carboxylic acids disclosed, particularly the *tetra* sodium salt of ethylene diamine tetra-acetic acid, are incompatible with alkali metal fatty acid soap compounds, the applicant is believed entitled to an allowance of the present claims which cover a detergent composition of such alkali metal fatty acid soap compounds in admixture with the new compounds of the instant invention which are compatible with such soap compounds, and in this respect, making an operable improvement over the Munz combination.

In view of the above remarks a prompt action on reconsideration of the final rejection of record is re-

spectfully solicited along with entry of the present amendment for the purposes of allowance or appeal. The amended claims are believed free of formal objection and patentable to the applicant.

Respectfully submitted.

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His Attorney.*

MARCH 18, 1949.
MNR:W.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Paper No. 13

Address only: The Commissioner of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the examiner in charge of this application. Lawrence C. Kingsland, Commissioner of Patents.

Division: 64. Applicant: Frederick C. Bersworth. Ser. No. 657,893. Filed March 28, 1946. For Washing Compositions.

MALCOLM N. RICH,
*11 Park Place, New York 7, New York.*

Applicant's remarks and the amendment filed March 19, 1949, have been carefully considered. The amendment however has not been entered since it does not place this case in condition for allowance.

The amendment proposes to cancel claim 19 and to amend claims 16 and 17.

The proposed amendment to claim 16, line 3, to insert—alkali metal fatty acid—before "soap" is regarded as new matter, no basis for which is found in the original disclosure. The proposed amendments to claims 16 and 17 would also not overcome the rejection thereof on the ground of insufficient disclosure as set forth in the last Office action.

The rejection of the claims as set forth in the last Office action is regarded as proper and is adhered to.

The proposed amendment will be entered for purposes of appeal when an appeal is filed, subject to the proviso that the Ex parte Mevey practice does not obtain.

A proper response to the last Office action must be made by July 28, 1949 to avoid the holding of abandonment.

P. M. NASH,
*Examiner.*

JG/mk.

In the United States Patent Office

Applicant: Frederick C. Bersworth. Invention: Washing Compositions. Division #64. Serial No. 657,893. Filed: March 28, 1946.

REQUEST FOR RECONSIDERATION

Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: The Examiner is respectfully requested to reconsider his rejection of claims 16 to 18, inclusive, as amended in A(4) filed March 19, 1949, on the grounds of "new matter" and "inadequate disclosure" in view of the following remarks:—

## Remarks

The Examiner has apparently overlooked the fact that applicant by the *inclusion* of Munz patent 2240957 in his specification (Page 2, paragraph 3) has thereby made the *entire* disclosure of this Munz patent a part of his disclosure and is accordingly *entitled* under recognized principles of patent law to amplify his disclosure by any part or all of the disclosure of this patent *without* objection thereto as "new matter". The Patent Office has repeatedly upheld this rule of patent law as the Examiner can easily ascertain.

In view of this basic principle of patent law, applicant is fully entitled to amplify his disclosure or to limit his claims to alkali metal fatty acid soap compounds as proposed in amendment (4) where the *disclosure* of the Munz patent mentioned by applicant in his specification is relied on for such identification of "soap" compounds, as pointed out by applicant in the remarks of the previous amendment (4) *on page 4 the first full paragraph.*

That this identification of "soap" compounds is *not contrary* to the clear intent and meaning of applicant in his specification has also been pointed out in the remarks of amendment (4).

That this identification of "soap" compounds is *not contrary* to the accepted meaning is evidenced by reference to standard chemical dictionaries of which the following are made of record and referred to:

Hackh's (Grant's Edition) (3rd Edition), page 777, identifies the word "soap" as:

"soap: a salt of one or more of the higher fatty acids with an alkali or a metal."

Van Nostrands' scientific Encyclopedia (1938 Edition), page 434, col. 1, last paragraph, also identifies the alkali metal salts of the fatty acids as "soaps".

The Condensed Chemical Dictionary (3rd Edition) on page 577 identifies "soap":

"Usually the sodium salt of oleic, stearic or palmitic acids or a mixture of these."

In view of the above-cited authorities it is believed self-evident that the term "soap" is correctly used by applicant in his specification and in conformity with the disclosure of the Munz patent mentioned in applicant's specification applicant is entitled to amplify his disclosure to more particularly identify "soap" as consisting of the alkali metal salts of the fatty acids or as "alkali metal fatty acid soap compounds" as now recited by claims 16 to 18.

Further, it is submitted that in view of the above remarks and cited authorities, the Examiner's rejection on "inadequate disclosure" is completely traversed. The term "soap" as used by applicant clearly meaning, in view of Munz and these cited authorities, the alkali metal fatty acids soap compounds well known and recognized in the art.

It is further urged that as all of these "soap" compounds are known and recognized in the art as being water soluble, the Examiner's rejection on this point is unwarranted as no specific example of such a water soluble soap compound is required.

In view of the above remarks a favorable action on reconsideration is requested so that this case may be passed promptly to issue with allowed claims 16 to 18, inclusive.

Respectfully submitted.

FREDERICK C. BERSWORTH,
By MALCOLM N. RICE,
*His Attorney.*

JULY 12, 1949.
MNR:W.

In the United States Patent Office

Applicant: Frederick C. Bersworth. Invention: Washing Compositions. Division No. 64. Serial No. 657,893. Filed March 28, 1946.

APPEAL TO THE BOARD OF APPEALS

Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: I hereby appeal to the Board of Appeals from the decision of the Principal Examiner in finally rejecting claims 16 to 18, inclusive, of the above identified application.

The following are the points of the decision on which the appeal is taken:

1. The Principal Examiner erred in finally rejecting the claims upon the references of record.
2. The Principal Examiner erred in finally rejecting the claims for the reasons of record.
3. The Principal Examiner erred in not allowing any or all of these claims.

The appeal fee of $15.00 is enclosed herewith. An oral hearing is requested at the convenience of the Board.

Respectfully,

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His Attorney.*

JULY 27, 1949.
MNR:W.

In the United States Patent Office

Applicant: Frederick C. Bersworth. Invention: Washing Compositions. Division: #64. Serial No. 657,893. Filed: Mar. 28, 1946.

AMENDMENT (5)

Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: Please amend the above application as follows:

## CLAIMS

Claim 16, line 3, erase "compound"; lines 5 and 7, change "alkalene" to—alkylene—; line 5 (from bottom) after "group" insert —consisting of —.

Claim 18, lines 2 and 3 (from bottom) erase "groups" insert—group consisting of—.

## REMARKS

The above amendments are for the purpose of putting these claims in condition for allowance by correcting the wording and spelling in the places specified in accordance with the Examiner's suggestion. The Examiner's courtesy in calling attention to these necessary corrections is gratefully acknowledged.

Prompt allowance of this application is solicited.

Respectfully submitted,

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His Attorney.*

December 30, 1949.
MNR:W.

## REFERENCE CITED

The following references are of record in the patented file of this patent:

UNITED STATES PATENTS

| *Name* | *Number* | *Date* |
|---|---|---|
| Bersworth | 2,387,976 | Oct. 30, 1945 |
| Munz | 2,240,957 | May 6, 1941 |
| Bersworth | 2,407,645 | Sept. 17, 1946 |
| Bersworth | 2,412,945 | Dec. 24, 1946 |

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Pol–85

Address only the Commissioner of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

MALCOLM N. RICH,
*11 Park Place, New York 7, New York.*

MARCH seventh, 1950.

*Notice of allowance*

The application for patent identified below has been examined and found allowable for issuance of Letters Patent. Applicant: Frederick C. Bersworth. Title of invention or improvement: Washing Compositions.

| *Serial No.* | *Filing Date* | *Number claims allowed* |
|---|---|---|
| 657,803 | March 28, 1946 | 3 |

With the allowance of the application the final fee becomes due. This fee is thirty dollars ($30) plus one dollar ($1) for each claim allowed in excess of twenty (20) and must be paid within 6 months from the date of this notice. Failure to remit the final fee will result in the patent being withheld from issue. In remitting this fee identify the application to which it applies by giving the following information: Name of inventor, title of invention, serial number, date of filing, date of allowance, and if assigned, the names of the assignees. The final fee will not be received from anyone other than the applicant, his assignee or attorney, or a party in interest as shown by the records of the Patent Office.

405365—56——13

If it is desired to have the patent issued to an assignee or assignees an assignment containing a request to that effect, together with the fee for recording the same, must be filed in this Office on or before the date of payment of the final fee.

The patent will be issued and forwarded approximately 4 weeks after receipt of the final fee.

By direction of the Commisisoner.

P. M. NASH,
*Examiner.*

*Final fee paid to the Commissioner of Patents*

AUGUST 31, 1950.

#422. (Be careful to give correct Serial No.) Serial No. 657,893. Inventor: Frederick C. Bersworth. Patent to be issued to: Frederick C. Bersworth. Name of invention, as allowed: Washington Compositions. Date of paymnt:——. Fee: $30.00. Date of fiing: March 28, 1946. Date of circular of allowance: March 7, 1950.

The Commissioner of Patents will please apply the accompanying fee as indicated above.

——— ———,
*Attorney.*

Send patent to Malcolm N. Rich, 11 Park Place, New York 7, N. Y.

Final fees will not be received from other than the applicant, his assignee or attorney, or a party in interest as shown by the records of the Patent Office, nor will they be applied in pending applications.

NUMBER (Series of 1935)

657893

DIV. 67

PATENT NO. 2524218

1946

DATED OCT 3 1950

(EX'R'S BOOK) 14-11

Name FREDERICK C. BERSWORTH,

of VERONA,

State of NEW JERSEY

Invention ~~ALKYLENE POLYAMINE DERIVATIVES~~ Washing Composition

ORIGINAL

APPLICATION FILED COMPLETE MAR 28, 1946

PARTS OF APPLICATION FILED

Petition, Specification, Oath, First Fee $30 sheets ~~Drawing~~, MAR 28, 1946

Division of App., No. ....., filed ....., 19....

Examined and passed for Issue [illegible], 19[illegible] Exr. Div. [illegible]

Notice of Allowance ....., 19..

By Commissioner.

Final Fee Rec'd 30 Sept 1950, 19..

Attorney ~~FRANK N. HOUGHTON 35 EAST ST ELMWOOD MASS~~

Malcolm N. Rich 11 Park Place, Newark 2, N. J.

Associate Attorney .....

No. of Claims Allowed 3 Print Claim 1 in O. G. Class 252-11.6

Title as allowed Washing Composition

10—11978-1

# 1946

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8. REVOCATION POWER OF ATTORNEY May 2-48
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*Plaintiffs' Exhibit No. 7, C. A. 1637- 4*

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE

Filed September 18, 1956. Harry N. Hull, Clerk.
*To all persons to whom these presents shall come, Greeting:*

This is to certify that the annexed is a true copy from the records of this office of the File Wrapper and Contents of the Application of Frederick C. Bersworth, filed July 17, 1948, Serial Number 39,377, for Hydroxy Derivatives of Alkylene Mono- and Poly-Amines and Method of Forming Same.

In testimony whereof I have hereunto set my hand and caused the seal of the Patent Office to be affixed at the City of Washington, this seventeenth day of February, in the year of our Lord one thousand nine hundred and fifty-six and of the Independence of the United States of America the one hundred and eightieth.

ROBERT C. WATSON,
*Commissioner of Patents.*

Attest:

C. G. RAINS,
*Certifying Officer.*

NUMBER (Series of 1948)

39377

DIV. 6

PATENT NO.

1948

DATED

(EX'R'S BOOK) 44-108-61

Name FREDERICK C. BERGSMITH

Pat 2352

SC A 7 Z

of VERONA

State of NEW JERSEY

Invention HYDROXY DERIVATIVES OF ALKYLENE MONO - AND POLY-AMINES AND METHOD OF FORMING SAME

ORIGINAL

APPLICATION FILED COMPLETE JULY 17, 1948

PARTS OF APPLICATION FILED

Petition, Specification, Oath, First Fee $30 — JULY 17, 1948

~~drawings~~

Examined and passed for Issue , 19 Ex. Div.

Notice of Allowance , 19

By Commissioner

Final Fee , 19

Attorney MALCOLM N. RICH 11 PARK PLACE NEW YORK 7, N. Y.

Associate Attorney

No. of Claims Allowed Print Claims in O. G. Class

Title as allowed

Filed , 19

Division of App., No.

MALCOLM N. RICH
PATENT ATTORNEY
11 PARK PLACE, NEW YORK 7, N. Y.,

*BArclay 7–6066, July 16, 1948.*

Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: Re: Appln. of Frederick C. Bersworth "Hydroxy Derivatives of Alkylene Mono- and Poly-Amines and Method of Forming Same"

I send herewith for filing an application for patent in this name. The enclosures are:—

1. Petition, Specification, 9 Claims and Oath;
2. Check for $30.00 Filing Fee.

Respectfully,

MALCOLM N. RICH.

MNR:W.
Encl.

APPLICATION FOR UNITED STATES LETTERS PATENT
PETITION WITH POWER OF ATTORNEY

Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: Your Petitioner, Frederick C. Bersworth, a Citizen of the United States, and a resident of Verona, County of Essex, and State of New Jersey, and whose Post Office address is 46 Marion Road, Verona, New Jersey; prays that Letters Patent of the United States may be granted to him for Improvements in Hydroxy Derivatives of Alkylene Mono- and Poly-Amines and Method of Forming Same set forth in the annexed specification.

And he hereby appoints Malcolm N. Rich, of 11 Park Place, New York City 7, New York (Reg. No. 13,142), his attorney, with full power of substitution

and revocation, to prosecute this application, to make alterations and amendments therein, to sign the drawings, to receive the Letters Patent when issued, and to transact all business in the Patent Office connected therewith.

Signed at Framingham, Massachusetts, this 13th day of July, 1948.

Frederick C. Bersworth.
FREDERICK C. BERSWORTH.

SPECIFICATION

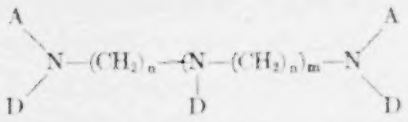
*To All Whom It May Concern:*

Be it known that I, Frederick C. Bersworth, a Citizen of the United States, and a resident of Verona, County of Essex, and State of New Jersey, have invented certain new and useful Improvements in Hydroxy Derivatives of Alkylene Mono- and Poly-amines and Method of Forming Same of which the following is a specification.

This invention relates to derivatives of alkylene mono- and poly-amines and more particularly to the mono- and poly-hydroxy derivatives of said alkylene mono- and poly-amines which derivatives are of wide utility in the art of chemistry and of chemical processes wherein compounds having strong detergent and wetting properties which are compatible with water soluble soap compounds and which in aqueous solutions form highly stable complexes with metal ions present in the solution, are desirably employed.

This application is a continuation-in-part application of my application Serial No. 657,893, filed March 28, 1946, in which application was disclosed and

claimed alkylene poly-amine derivatives corresponding to the following structural formula:



wherein n represents one of the numerals 2 and 3; m has a value selected from the group consisting of zero (0), 1, and any multiple of 1; D is one of the substituent groups consisting of $-CH_2 \cdot COOH$, $-CH_2 \cdot CH_2 \cdot COOH$, and a basic metal salt thereof; and wherein A is a hydrocarbon group containing at least two (2) carbon atoms and having not more than two (2) hydrogen atoms thereof replaced by one of the substituent groups consisting of $-CH_2 \cdot COOR$, $-(CH_2)_2 \cdot COOR$, $-CH_2 \cdot COOM$, $-(CH_2)_2 \cdot COOM$, $-SO_3H$, $-SO_3M$, $-OR$, and $-OR'OR$ wherein R is alkyl or hydrogen, M is an alkali metal and R′ is alkylene, said groups being spaced at least two carbon atoms from the nitrogen atoms.

The present invention relates to compounds conforming to this general formula in which A consists of a mono- or poly-hydroxy hydrocarbon group, the compound otherwise conforming to the formula above given.

I have discovered that these mono- and poly-hydroxy derivatives of alkylene mono- and poly-amines are highly effective sequestering agents in aqueous solutions for metal ions present therein and are particularly effective in sequestering polyvalent metal ions having a valency of at least three (3) forming complexes therewith which resist decomposition and precipitation of the metal ions by normal precipitating agents therefor over a wide range of temperatures and over a wide range of pH; and that

said complexes are compatible and non-reactive with water-soluble soap compounds present in the solution.

The acidic hydroxy derivative compounds of the present invention are best formed by carboxylating a mono- or poly-hydroxy amine in accordance with the method described and claimed in my prior patent No. 2,407,645 issued September 17, 1946, acidifying the reaction solution to neutralize the alkali metal salt thereof and to precipitate the hydroxy-amino acid therefrom, concentrating the solution to small volume to recover additional amounts of the acid, and separating the precipitated amino acid product from the solution. Alternatively, the alkali metal salt of the hydroxy-amino acid present in the reaction solution may be recovered therefrom by evaporating off the major portion of the water of hydration to form a concentrated solution of the salt and dehydrating the salt preferably under vacuum. This salt upon resolution in water and on neutralization with a strong mineral acid precipitates the hydroxy-acid as before described.

As an example of this method of producing these hydroxy-derivatives of alkylene poly-amines, the following example is given:

One (1) molar weight of mono-ethanol-amine is processed, in accordance with the method of my above identified patent, with two (2) moles of sodium cyanide and two (2) moles of formaldehyde, at a reaction temperature approximating 85° C, using a catalytic amount of caustic soda (NaOH) in the reaction solution to maintain a pH of about 9. The reaction product obtained is the sodium salt of ethanol-amine di-acetic acid.

This amino acid salt is recovered from the solution by evaporating the solution to a syrupy consistency and dehydrating the solution under vaccum.

On re-dissolving this salt in water to form a highly concentrated solution thereof and neutralizing the same with a strong mineral acid ($H_2SO_4$ or HCl) using a sufficient excess of the acid to provide a pH of about 6.5 in the solution, the anhydride of ethanol amine di-carboxylic acid is precipitated in a form readily separated by filtration from the supernatant liquor.

As another example, mono- and di-ethanol ethylene diamine, similarly processed, will yield the mono-ethanol-thylene diamine-triacetic acid salt and the di-ethanol-ethylene diamine-di-acetic acid salt, respectively, which may be recovered from the reaction solution as the salt or as the anyhydride compound in the manner hereinabove described.

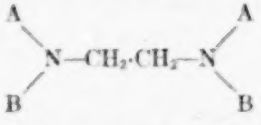
Mono- and poly-ethanol amines and poly-amines containing at least one displaceable amino hydrogen, but preferably more than one said amino hydrogen, are generally subject to carboxy-methylation substitution in accordance with the process of my above identified patent to form mono- and poly-corboxylic (acetic) acid salts analogous to those identified above.

Similar and substantially equivalent chemical compounds are obtained on carboxy-methylation of mono- and poly-propanol amines and poly-amines.

For the purposes of the present invention, however, I have found that while all of these ethanol and propanol derivatives of mono- and poly-amines are chemically equivalent compounds, the effectiveness and efficiency of those compounds as complex-forming agents towards metal ions in solution therewith varies with variation in composition.

Of all of these compounds I have found that the most stable and reliable compounds for general purpose use especially in the presence of water-soluble soap compounds are those derivatives of ethylene

diamine wherein two (2) of the amino hydrogens of the diamine have been displaced by ethanol or propanol and the other two (2) of the amino hydrogens have been displaced by acetic acid groups. Such a compound is indicated by the following formula:




wherein A represents ethanol ($-CH_2 \cdot CH_2 \cdot CH$) or propanol ($-CH_2 \cdot CH_2 \cdot CH_2 \cdot OH$) and B represents an acetic acid group ($-CH_2 \cdot OOH$) wherein the H may be displaced by substantially any basic metal ion but preferably is displaced by a strongly basic metal ion such as an alkali or alkaline earth metal ion.

This compound is formed by carboxy-methylating di-ethanol or di-propanol ethylene diamine in accordance with the process of my prior patent, above identified, to displace the remaining two (2) amino hydrogens of the diamine with acetic acid groups ($-CH_2 \cdot COONa$). Where the process is properly conducted the reaction is substantially quantitative where the materials entering into the process are substantially pure. On a commercial basis yields approximating 90 to 95% of the theoretical may be consistently obtained.

In commercial practice I prefer to first recover the alkali metal salt of this di-ethanol (or di-propanol) ethylene diamine di-acetic acid from the reaction solution by evaporating off the major portion of the water of solution until the solution has a syrupy consistency and careful drying or dehydrating under vacuum.

The purified salt is then re-dissolved in water to form a 20–30% solution and the anhydride thereof is obtained by acidifying the solution with a strong min-

eral acid ($H_2SO_4$ or HCl) to a pH of about 6.5 at which the anhydride is the least soluble.

As an example of the effectiveness and utility of the di-ethanol ethylene diamine di-acetic acid compound as a sequestering agent for polyvalent metal ions in aqueous solutions therewith, the following test results are given:

As is well known in the art most commercial waters contain at least a small amount of iron. This iron will precipitate out of solution when the pH of the solution is made alkaline. In the many chemical processes conducted in alkaline pH solutions it is often desired to inhibit and prevent the precipitation of such basic iron compounds. Typical of these chemical processes is that involving the washing or laundering of organic fibrous materials wherein a water-soluble soap, such as an alkali metal-fatty acid soap compound, is employed. In such operation the iron content of commercial water is generally precipitated either in the form of ferric hydroxide or as an insoluble iron-fatty acid soap compound which compounds usually become affixed to the surface of the washed or laundered material requiring special acid treatment to be removed therefrom.

In the case of one sample of commercial water containing, by chemical analysis, 4 parts per million of Fe * * *, the addition of sodium hydroxide in an amount providing a 3% sodium hydroxide solution with heating to the boiling point was sufficient to precipitate the iron content of the solution as ferric hydroxide ($Fe(OH)_3$). The addition of a small amount of an alkali metal—fatty acid soap to the solution, in substitution for the sodium hydroxide, with heating was sufficient to form a precipitate of the insoluble iron-fatty acid soap.

By the addition, to the commercial water, of an amount of di-ethanol ethylene diamine di-acetic acid providing approximately one molar weight of the acid per molar weight of the iron, calculated as Fe, the precipitation of $Fe(OH)_3$ and the formation of insoluble iron-fatty acid soap compounds is completely inhibited, even in the presence of sufficient free caustic alkali to increase the pH to as high as 12 and on extended heating at the boiling point of the solution or on several days standing.

Similar test results were obtained in water containing various other metals ions commonly found in commercial waters and the complexes formed by all of these metal ions with di-ethanol ethylene diamine di-acetic acid or the alkali metal salts thereof, appear completely stabilized against precipitation by the usual precipitating agents therefor over a wide range of pH and over a wide range of relative concentrations of precipitant to complex.

The exact composition of the complex formed with these metal ions by di-ethanol ethylene diamine di-acetic acid is the subject of conjecture but it is believed that these metal ions must be held in un-ionized complex combination by the residual valency bonds of the amino nitrogens set free and made active upon solution and ionization of the ethanol amino acid in water.

Where the hydroxy-poly-amino acid contains but one ethanol substituent group and three acetic acid substituent groups, such as is present in mono-ethanol ethylene diamine tri-acetic acid, the effectiveness of the compound towards some polyvalent metal ions is somewhat different. In the case of iron, for example, the complex formed appears to be primarily one holding the iron in its ferric state whereas the di-ethanol

amino acid appears to hold the iron in its ferrous state.

Di-propanol ethylene diamine di-acetic acid and mono-propanol ethylene diamine tri-acetic acid react substantially identically to that above described in connection with the di-ethanol and mono-ethanol amino acids.

Similarly, other aliphatic amines and poly-amines form ethanol and propanol acetic acid derivatives which are substantial equivalents to those hereinabove described, insofar as their complex-forming properties with metal ions in aqueous solutions may be concerned.

One of the major advantages of the complex-forming compounds of the present invention, over those heretofore employed in the art, is that the compounds themselves and the metal complexes thereof are soluble in water over a wide range of pH, from strongly acid to strongly alkaline, due to the presence therein of the non-ionic solubilizing ethanol and propanol groups, and due to the fact that the weakly ionic carboxylic groups upon complex-formation become substantially non-ionic.

Many of these amino acids may be recovered from concentrated aqueous solutions as anhydrides upon acidification with a strong mineral acid to a pH at which de-hydration of the amino acid occurs. Other means for de-hydrating the amino acid may be employed to recover same from aqueous solution. However, in the usual small amounts of these amino acids required to sequester the metal ions present in most commercial waters the presence of relatively large amounts of free acid or free alkali in the solution is ineffective in causing decomposition of the hydroxy-amino acids, their salts or the metal complexes thereof.

Having hereinabove generically and specifically disclosed the present invention and having given several specific examples of the same and indicated the extent of utility thereof in one field of use, it is believed apparent that the invention may be widely modified without essential departure therefrom and all such modifications and departures are contemplated as may fall within the scope of the following claims:

What I claim is:

5. The chemical compound di-ethanol ethylene diamine diacetic acid, the basic metal salts thereof, and the metal ion complexes thereof.

6. The chemical compound di-propanol ethylene diamine diacetic acid, the basic metal salts thereof, and the metal ion complexes thereof.

8. The method of producing hydroxy alkylene mono- and poly-amine derivatives which comprises treating a hydroxy aliphatic amine containing at least one displaceable amino hydrogen with sodium cyanide and formaldehyde in an alkaline pH solution at a temperature approximating 85° C to displace the amino hydrogens of the amine with acetic acid groups, removing the major portion of the water of solution and dehydrating the salt under vacuum.

9. The method of claim 8, wherein the separated salt product is redissolved in a small amount of water and an amount of a strong mineral acid is added to the solution to neutralize the said salt and to provide an excess acid sufficient to insolubilize the hydroxy-amino acid as an anhydride.

In witness whereof, I have hereunto signed my name.

Frederick C. Bersworth.

FREDERICK C. BERSWORTH.

*Oath*

STATE OF MASSACHUSETTS,

*County of Middlesex, ss:*

Frederick C. Bersworth, the above-named Petitioner, being duly sworn, deposes and says that he is a Citizen of the United States, and a resident of Verona, County of Essex, and State of New Jersey; that he verily believes himself to be the original, first and sole inventor of the Improvements in Hydroxy Derivatives of Alkylene Mono- and Polyamines and Method of Forming Same described and claimed in the annexed specification; that he does not know and does not believe that the same was ever known or used before his invention or discovery thereof, or patented or described in any printed publication in any country before his invention or discovery thereof, or more than one year prior to this application; that, as to the portions of his invention that are common to this application and of his prior filed application Serial No. 657,893, filed March 28, 1946, identified in this application, he does not knew and does not believe that the same were in public use or on sale in the United States for more than one year before the respective filing date of said prior filed application; that as to the portions of this invention that are described and claimed in the above identified application which were not disclosed in said prior filed application, he does not know and does not believe that the same were in public use or on sale in the United States for more than one year prior to this application; that said invention has not been patented in any country foreign to the United States on an application filed by him or his legal representatives or assigns more than twelve months prior to this application; and that no application for patent on said

improvements has been filed by him or his legal representatives or assigns in any country foreign to the United States.

Frederick C. Bersworth.
FREDERICK C. BERSWORTH.

Sworn to and subscribed before me this 13th day of July 1948.

[SEAL] MARY A. COLEMAN,
*Notary Public.*

My commission expires November 29, 1954.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Paper No. 2

Address only the Commissioner of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the Examiner in charge of this application. Lawrence C. Kingsland, Commissioner of Patents.

Division: 6—Room 7616 Mexes. Applicant: Frederick C. Bersworth. Ser. No. 39,377. Filed July 17, 1948. For hydroxy derivatives of alkylene monopolyamines and method of forming same.

MALCOLM N. RICH,
*11 Park Place, New York 7, New York.*

This application has been examined.

References cited:

Bersworth—2,407,845, Sept. 17, 1946, 260–534.
Bersworth—2,413,856, Jan. 7, 1947, 260–534.
Bersworth—2,428,353, Oct. 7, 1947, 260–534.
Johnson (Br.)—388,874, Mar. 9, 1933, 260–534.

Claim 8 is rejected as being unpatentable over the above cited patent No. 2,407,645 or the British patent. The process is old and conventional. No invention is perceived.

Claims 3 and 5 appear to be identical and one should be cancelled.

Claims 1, 2, 5, 6 and 7 are rejected as involving Markush type claims of varying scope. This is not permissible and should be corrected.

Claims 1, 2, 5, 6 and 7 are rejected as being alternative. The conventional term is—*a member of the group consisting of* ——. The conventional term should be inserted appropriately in these claims.

Claim 4 is rejected as being unpatentable over the Bersworth patent No. 2,407,645 which discloses the compound (Col. 5). There apparently is no continuity of disclosure with this patent. Hence the patent is a bar and the problems of dedication to the public and laches are raised.

Claims 1 to 7 inclusive are rejected as being unpatentable over either of the Bersworth patents cited above, particularly No. 2,413,856 which discloses the class of compounds covered as well as it is disclosed in this application. Since applicant's parent case apparently does not have an adequate disclosure to form a basis of the claims, and since that disclosure is not common with the patent disclosure, the patents apparently amount to a bar and raise the problem of dedication to the public as well as *laches* in claiming.

Claims 1 to 7 inclusive are rejected as being based on an inadequate disclosure. All the classes of compounds covered are not based on representative complete working examples, which is necessary to carry out the provisions of Sec. 4888 R. S. (Ex parte Fischer, File of U. S. Patent No. 2,434,357; Ex parte Coleman et al., File of U. S. Patent No. 2,417,809.

No generic claim having been allowed, tentative election is required as follows:

(I) To the compounds of claim 4

(II) To the compounds of claim 5 (claim 3)

(III) To the compounds of claim 6

(IV) To the anhydrides of claim 9 (30 J. P. O. S. 550–552)

Applicant is advised that his response to be complete under Order 3527 must include an election of species, or a statement that in his opinion all claims are in fact readable on a designated species, which applicant must identify, and that such statement will be construed as an election under the Order.

J. H. Byer,
*Acting Examiner.*

In the United States Patent Office

Applicant: Frederick C. Bersworth. Invention: Hydroxy Derivatives of Alkylene Mono- and Poly-Amines and Method of Forming Same. Examiner's Room 7616. Division #6. Serial No. 39,377. Filed: July 17, 1948.

Amendment (1)

Hon. Commissioner of Patents,
*Washington 25, D. C.*

Sir: In response to the Official Action of October 28, 1948, please amend the above application as follows:

*Claims*

Claim 1, line 3, erase "represents one" insert—is a member of the group consisting—; same line, erase "represents" (second occurrence); line 4, after "value" insert—selected—; line 5, before "one" in-

sert—a member of—; line 8, erase "derived" insert —selected—.

Claim 7, line 5, before "hydrogen" insert—selected from the group consisting of—.

*Remarks*

Claims 1 and 7 (along with depending claims 2 to 4) have been amended to overcome the Examiner's objection to the formal wording of the claims.

Reconsideration of the patentability of these claims and of the unamended claims is respectfully requested in the light of the following argument:

Claims 1, 2, 5, 6 and 7 are rejected as involving Markush type claims of varying scope. The Examiner's attention is directed to the fact that this ground for rejection applies *only* to claims wherein the applicant after establishing his Markush grouping of a plurality of equivalents (for which no known classification appears) then attempts to sub-divide these plurality of equivalents into sub-genus divisions containing *more than one (1) of these members of the Markush group.*

Where only one (1) member of the Markush group is specified then the claim assumes the status of a species claim. Applying this to claims 2, 3 and 4, it is to be noted that in limiting D to acetic acid group a species claim is defined irrespective of the plurality of members specified in the A group as these members are species monohydroxy hydrocarbon compounds.

Claim 3 is specific to ethylene diamine, one of the members of the Markush group amines.

Claim 4 is still more specific than claim 3.

Claims 5 and 6 merely define species compounds chemically instead of structurally and are believed al-

lowable under the general doctrine that the applicant is permitted claims of different wording defining substantially the same invention in the same application.

Claim 7 is clearly species to claim 1 defining a species hydroxy group for A and a species D group.

The examiner rejects claims 4 and 8 and claims 1 to 7 on Bersworth patent 2,407,645 or British and on Bersworth 2,413,856, respectively. Reconsideration is requested.

The present application is a continuation-in-part application of application Serial No. 657,893 filed *March 25, 1946*, which is copending herewith. The filing date of this parent application is *prior* to the issue date of Patent 2,407,645 (September 17, 1946). The parent application having been filed prior to the issue date of the patent, the patent is not available as a reference against the parent patent or against the present continuation-in-part application. As to any matter disclosed but not claimed in the patent, dedication to the public is expressly negatived by the filing of the instant parent and continuation-in-part applications.

In view thereof, withdrawal of Patent 2,407,645 as a reference against the instant invention is requested.

Substantially the same objection to Patent 2,413,856 as a valid reference is also made. This patent has an issue date of January 7, 1947. The parent application having been copending with this patent, the patent is not available as an anticipation of the invention therein disclosed or by the invention divided out and claimed in the present application. Dedication to the public is expressly negatived by the chain of parent and continuation applications involved. Withdrawal of this patent also is requested.

The same objections are made to Patent 2,428,353 and withdrawal of the same as a reference is requested.

In refutation of the Examiner's statements, top of page 2, of the Action, regarding the extent of disclosure in the parent application the Examiner's attention is directed to the disclosure page 2 of the application and to generic claim one of the pending parent application. The substituent group —OR'.OR conforms to the species of the instant application.

The rejection of claims 1 to 7 on inadequate disclosure is believed unsound and improper and reconsideration is requested. The specific example given on page 3; the other specific examples given on top of page 4 down to line 15; the explanation given in paragraph 4 on page 4; and the disclosure pages 6 and 7 as to utility of one species and the disclosure page 8, paragraphs 2 and 3 as to equivalent species, is believed clearly ample to support genus and species claims.

The Examiner is believed in error by reason of the fact that in the general rule stated he has overlooked the significant exception to the rule which is to the effect *the rule only applies where the chemical reaction involved is obscure or uncertain.* The Examiner certainly cannot contend that the carboxymethylation process is obscure or uncertain as to all mono- or poly-hydroxy amines. In the absence of this contention the Examiner's rejection is of no merit.

The Examiner moreover has not made of record any reference which discloses that one or more of the compounds covered by the generic claim cannot or does not react as stated by the applicant. In the absence of this evidence the Examiner's rejection

on possible inoperativeness is without merit as being just a guess or personal opinion.

Pending the allowance of a generic claim applicant elects to prosecute claims to the species of the specific example on page 3 and of claims 3 and 5.

A more favorable action on reconsideration is respectively solicited.

Respectfully submitted.

FREDERICK C. BERSWORTH,

By ------ ------,

*His Attorney.*

MARCH 18, 1949.
MNR: W.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Address only the Commissioner of Patents, Washington 25, D. C.

Paper No. 4

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

IV-Mc-jek

Please find below a communication from the EXAMINER in charge of this application. Lawrence C. Kingsland, Commissioner of Patents.

Division: 6—Room 7616. Applicant: Frederick C. Bersworth. Ser. No. 39,377. Filed July 17, 1948 For Hydroxy Derivatives of Alkylene Mono and Poly-Amines and Method of Forming Same.

MALCOLM N. RICH,
*11 Park Place, New York 7, N. Y.*

Responsive to amendment filed March 19, 1949.

Additional references made of record:

Ulrich et al.—1,972,465, Sept. 4, 1934, 260–465.5.
Hoffman et al.—2,017,537, Oct. 15, 1935, 260–534.
Kiprianow, Beilstein (Handbuch, 4th Ed.) 2nd Suppl., vol. 4, p. 801 (1942).
Copy in Sci. Library.

Claims 1 to 9 inclusive are rejected as being unpatentable over the Beilstein reference cited above in view of the Ulrich et al. or Hoffman et al. patents, both cited above. Applicant has presented only one working example in the specification, although this is incomplete (Rule 71b). This example produces the compound of the Beilstein reference. Applicant's method of producing this compound as well as the acetic acid compounds claimed is old and conventional, as disclosed by Ulrich et al. and as admitted by the applicant (Remarks of March 19, 1949, page 4). The Hoffman et al. patent discloses a process for preparation of the propionic acid compounds. If applicant's disclosure is adequate to support the claims, then by the same reasoning the art applied amounts to references for the claims. The process is of course old as disclosed by Ulrich et al. and admitted by applicant. Applying it to a particular raw material of the same class is non-inventive.

Claim 2 is again rejected as being alternative. The conventional term is—*a member of the group consisting of*—.

Claim 4 is again rejected as being unpatentable over the Bersworth patent No. 2,407,645 for reasons of record.

Claim 4 is rejected as being inept and ambiguous in that it covers a tricarboxylic acid, yet depends on claim 1 which is limited to dicarboxylic acids.

Claims 1, 2, 4, 5, 6, and 7 are again rejected as involving Markush groups of varying scope. The appli-

cant's attention is directed to *Ex parte Cook et al.* (76 U. S. P. Q. 196).

Claims 1 to 9 inclusive are again rejected as being unpatentable over either of the Bersworth patents of record for reasons of record in connection with claims 1 to 7. The applicant's remarks have been carefully considered but have not been found convincing as to the patentability of the claims.

Claims 1 to 7 inclusive are again rejected as being based on an inadequate disclosure. There should be a complete working example supporting each class of compound covered (Rule 71b; see citations of record; see also *Ex parte Haury*, File of U. S. Patent No. 2,477,872). While what applicant says in this regard might apply in some cases to process claims, it is not considered that the product claims are properly supported.

Claims 4, 6, and 9 are rejected as failing to read on the elected species.

There apparently should be cross-referencing between this application and applicant's S. N. 39,378, S. N. 737,725 and S. N. 735,018 (Rule 78a).

A patentable line of distinction should be maintained between this application and applicant's S. N. 39,378, S. N. 737,725 and S. N. 735,018. Where the claims conflict, applicant may be required to cancel the claims from all but one application (Rule 78b).

This application should be prepared for final action. No further amendments that do not place the case in condition for allowance or better form for appeal will be entered after final action unless accompanied by a proper showing of good and sufficient reasons why they were not earlier presented. Rule 116 and Commissioner's Notice of April 26, 1948.

*Examiner.*

In the United States Patent Office

Applicant: Frederick C. Bersworth. Invention: Hydroxy Derivatives of Alkylene Mono- and Poly-Amines and Method of Forming Same. Division: # 6. Room: # 7616. Ser. No. 39,377. Filed: July 17, 1948.

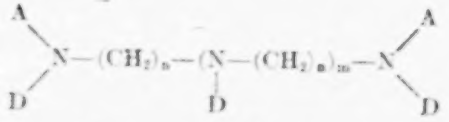
*Amendment (2)*

Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: In response to the Official Action of September 14, 1949, please amend the above application as follows:

*Claims:* Erase claims 1 to 4, inclusive, and claim 7, and substitute therefore the following claims:

10. The chemical compound conforming to the structural formula:



wherein n is a numeral selected from the group consisting of 2 and 3; m is a value selected from the group consisting of zero and a positive integer; D is a member of the substituent groups consisting of $-CH_2COOH$, $-CH_2-CH_2COOH$ and basic metal salts thereof; and A is a hydroxy substituent group selected from the group consisting of mono- poly-hydroxy hydrocarbon compounds.

11. The compound of claim 1, wherein D consists of $-CH_2COOH$ and A consists of ethanol.

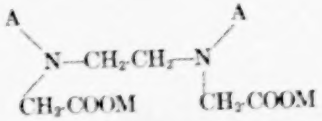
12. The compound of claim 1, wherein D consists of $-CH_2COOH$ and A consists of propanol.

13. The compound of claim 1, wherein the structure $-(CH_2)_n-(N-CH_2)_n)_m$ consists of

D

the ethylene group $-CH_2 \cdot CH_2-$, D consists of the group $-CH_2 \cdot COOH$, and A consists of the group $-CH_2 \cdot CH_2 \cdot OH$.

14. The chemical compound conforming to the structural formula:—




wherein A is a member of the group consisting of ethanol and propanol; and M is a member of the group consisting of hydrogen and a basic metal ion.

## Remarks

In response to the Official Action of September 14, 1949, rejected claims 1 to 4, inclusive and claim 7 have been erased and rewritten as new claims 10 to 14, inclusive, to avoid confusing amendments thereto in correcting same to overcome the Examiner's objections thereto.

Reconsideration of the rejection of retained product claims 5 and 6, and retained process claims 8 and 9, and an action on the new claims 10 to 14, inclusive, are requested in the light of the following remarks and argument.

I. In the rejection of claims 1 to 9, inclusive, on Beilstein in view of Ulrich or Hoffman, it is respectfully urged that the Examiner fails to recognize that the Beilstein reference discloses the compound

$$C_6H_{11}O_5N = HO \cdot CH_2 \cdot CH_2 \cdot N(CH_2 \cdot COOH).$$

This compound is identified by Beilstein as being an *oxy-ethylimino di-acetic acid.*

The compound *claimed* by applicant is an alkylene polyamine in which the amino hydrogens on the amino nitrogens have been displaced by the "A" and "D" groups specified.

This type of compound is *remote* from that disclosed by Beilstein and hence this reference is not a good basic reference unless the supplementary references relied on teach one skilled in the art to change the Beilstein compound to be that claimed.

It is believed obvious that neither Ulrich nor Hoffman so teach one skilled in the art to change or modify the Beilstein compound. Ulrich teaches the production of *nitriles* by causing *hydrocyanic acid* to act on the products obtainable by the reaction of primary or secondary hydroxyalkylamines with aliphatic carbonyl compounds (aldehydes and ketones). (See page 1, column 1, paragraph beginning line 9.) These hydroxyalkylamino carboxylic acids are of an entirely different structure than those herein claimed as the structural formulas given by Ulrich clearly indicate.

Hoffman discloses a method of producing hydroxyalkylamino carboxylic acids by reacting primary and secondary alkylol amines with *unsaturated carboxylic* acid compounds having an aliphatic carboxylic acid chain in which an unsaturated linkage between carbon atoms (olefinic linkage) is situated in the alpha, beta position to the carboxylic group. (See page 1, column 1, paragraph beginning line 7.)

The compounds produced by this process are clearly different in structure from those defined by applicant's claims as is self-evident from the structural formulas given in this reference.

It is accordingly urged that this combination of references fails to negative patentable novelty in the products claimed for the reason that neither Ulrich

nor Hoffman teaches how to convert the compound of Beilstein into the polyamino compound generically defined by claim 1 (now claim 10).

II. With reference to the Examiner's remarks in support of the rejection on these references, it is respectfully submitted that in the instant situation, the carboxymethylation reaction, per se, being old applicant is not required, under the general rule that that which is old is as delineated in the specification, to do more than to identify the process to be old and to tell on what compound the process is to be practiced to produce what product.

This applicant has done on page 3, lines 11 to 30, inclusive, in far greater detail than is required, and including the details of product recovery.

Again on page 5 the details of the process for forming the di-ethanol or di-propanol ethylene diamine diacetic acid compounds are given the recovery details.

In view of this disclosure the Examiner's remarks are believed unwarranted as to its completeness. It is sufficient to enable one skilled in the art to practice the method to produce the claimed product. This is all the patent statutes require in the way of a disclosure.

The Examiner's remarks lines 1 to 7 (from bottom) on page 1 of the Action are strongly objected to. There is no judicial justification for such an "if" basis of rejection. Anticipation is a question of fact. The references either do or do not disclose the same invention. Rejections based on conjecture are improper, per se.

III. Referring next to the Examiner's rejection on improper Markush grouping, the claims have been erased and rewritten to overcome this rejection. However, the Examiner's attention is directed to the

fact that the Commissioner of Patents recertly issued an order modifying this rule against Markush claims of varying scope and also permitted the joining of generic and Markush claims in one application.

IV. With reference to the rejection of product claims on the Bersworth patents. The mere fact that the process used is old, per se, does not negative novelty in product, as the two (process and product) are independent inventions. The statute itself differentiates between process and product.

In the instant case carboxymethylation is old. Applicant carboxymethylates di-ethanol ethylene diamine (for example) to form di-ethanol ethylene diamine di-acetic acid. This compound is new over any disclosed in the Bersworth patents as Bersworth discloses carboxymethylation of ethylene diamine *only*.

One skilled in the art does not know and cannot determine with that degree of certainty required for anticipation, whether or not the ethanol substituent groups on the ethylene diamine would be stable under the conditions of the carboxymethylation process. This fact remains open for determination. Determination of this fact; the production of the diethanol ethylene diamine di-acetic acid by this or by any other process; and the determination of its physical and chemical properties are also inventive acts on which patentable novelty of product are predicated. The Courts and Patent Office Tribunals have repeatedly ruled on this question and the authorities are all in favor of applicant on this question of patentable novelty.

V. Referring now to the rejection (page 2) on inadequate disclosure, the Examiner's attention is directed to the fact that the rule stated is not an absolute rule but one subject to many exceptions. The

primary exception is that which states that the rule does not apply when the chemical reaction involved is *obvious*. In the instant case the Examiner cannot contend that carboxymethylation of hydroxy amines generally cannot work substantially as stated by the applicant in his specification. The reaction once described with one hydroxyamine will obviously work in the same way with any other hydroxyamine. The only requirement involved is that there is at least one (1) amino hydrogen present in the amine for carboxymethylation substitution.

Therefore, it is submitted, the requirement for an example for *each* compound covered by the claims is clearly unwarranted and, as clearly, unnecessary under the statutes and under leading decisions on this point. The Examiner has cited a number of decisions none of which are in point. The better rule is that of *Ex parte Kropa* (P. O. Bd. Appl 63 U. S. P. Q. 16) wherein it was held to be *not absolutely* necessary to give specific examples as required by the Examiner.

Also see *Ex parte Sharma* (48 U. S. P. Q. 646) in support of applicant's contention as above given.

See also *Wayne* v. *Kingsland* (80 U. S. P. Q. 486) on the sufficiency of single disclosure in chemical cases to support generic claims. In this case the Court denied explicitly the existence of the absolute rule stated by the Examiner.

VI. With reference to method claims 8 and 9, the *additional* steps present in these claims over carboxymethylation, per se, are believed sufficient to support patentable novelty.

The comments of the Examiner regarding cross-referencing and line of distinction between copending applications has been noted and will be attended to

and observed, respectively, as the prosecution demands.

A favorable action on the retained and new claims is respectfully solicited.

Respectfully submitted.

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His Attorney.*

MARCH 6, 1950.
MNR: W

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

POL–90 Paper No. 6

Address only The Commissioner of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the EXAMINER in charge of this application. John A. Marzall, Commissioner of Patents.

Division: 6. Applicant: Frederick C. Bersworth. Ser. No. 39,377. Filed July 17, 1948. For Hydroxy Derivatives of Alkylene Mono- and Poly-Amines and Method of Forming Same.

MALCOLM N. RICH,
*11 Park Place, New York 7, New York.*

Responsive to amendment filed March 7, 1950.

Claims 11, 12 and 13 are rejected as depending on a cancelled claim. The claims should be complete as they would be printed.

Claims 10, 11, 12 and 14 are rejected as involving Markush groups of varying scope. This is not per-

missible (*Ex parte Cook et al.*, 76 U. S. P. Q. 196). The Commissioner's order referred to by applicant, while permitting a natural genus and a subgeneric Markush claim, is not considered to overcome or change the practice as stated in Ex parte Cook et al.

Claims 5, 10, 11, 13 and 14 are again rejected as as being unpatentable over the Beilstein reference of record, in view of the Ulrich et al. or Hoffman et al. patents of record. The applicant's arguments have been carefully considered but have not been found convincing as to the patentability of the claims.

Applicant has presented only one working example in the specification, although this is incomplete (Rule 71b). This example produces the compound of the Beilstein reference. Applicant's method of producing this compound as well as the acetic acid compounds claimed is old and conventional, as disclosed by Ulrich et al. and as admitted by the applicant (Remarks of March 19, 1949, page 4). The Hoffman et al. patent discloses a process for preparation of the propionic acid compounds. If applicant's disclosure is adequate to support the claims, then by the same reasoning the art applied amounts to references for the claims. The process is of course old and disclosed by Ulrich et al. and admitted by applicant. Applying it to a particular raw material of the same class is noninventive. Applicant continually refers to the conventionality of the *process*. However, these claims are *product* claims none of which are illustrated by complete examples of preparation or any characteristics. For that reason, and since applicant's deduction of the claimed *compounds* from an example old in the prior art, necessarily places this prior art compound in a class with the claimed compounds, no reason for excluding the Beilstein compound from

the Markush type claims is perceived. Hence, the claims are considered to be anticipated.

Claims 5, 10, 11, 13 and 14 are again rejected as being unpatentable over either of the Bersworth patents of record, particularly #2,413,856, which discloses the class of compounds covered as well as it it disclosed in this application. Applicant's parent application apparently does not have an adequate disclosure to form an adequate basis for the claims, the patents apparently amount to a bar, raise the problem of dedication or amount to *laches* in claiming.

Claims 5, 10, 11, 13 and 14 are rejected as being based on an inadequate disclosure. There should be, to constitute an adequate disclosure, a complete working example illustrating each *class* of compounds covered (Rule 71c; Ex parte Haury, File of U. S. patent #2,477,872, paper #14, 5 pp.-recon. paper #16, 2 pp.). While applicant's remarks might apply in some cases involving *process* claims, these product claims are considered to be lacking in adequate support.

Claims 6, 9, and 12 are rejected as failing to read on the elected species.

Applicant's remarks and the decisions cited have been carefully considered but have not been found convincing as to the allowability of the instant claims. Wayne v. Kingsland specifically refrains from establishing a general rule contra the patent Office practice and Rule 71b. The decisions cited by the Examiner are believed to represent the practice.

This action is made *final*.

H. SURLE,
*Examiner.*

In the United States Patent Office

Applicant: Frederick C. Bersworth. Division: No. 6. Room: No. 7616. Serial No. 39,377. Filed: July 17, 1948. Invention: Hydroxy Derivatives of Alkylene Mono- and Poly-Amines and Method of Forming Same.

Hon. Commissioner of Patents,
*Washington 25, D. C.*

Sir: Further prosecution of the above identified application is herewith abandoned in favor of application Serial No. 215,380, entitled "Hydroxy Derivatives of Alkylene Polyamines" filed March 13, 1951.

Respectfully,

Frederick C. Bersworth.
Frederick C. Bersworth.

March 12, 1951.
MNR:W.

Department of Commerce
United States Patent Office
Washington

POL-96 Paper No. 8

Address only the Commissioner of Patents, Washington 25, D. C.

All communication respecting this application should give the serial number, date of filing, and name of applicant.

Please find below a communication from the EXAMINER in charge of this application. John A. Marzall, Commissioner of Patents.

Division: 6—Room 7725. Applicant: Frederick C. Bersworth. Ser. No. 39,377. Filed: July 17, 1948.

For Hydroxy Derivatives of Alkylene Mono- and Poly-Amines and Method of Forming Same.

MALCOLM N. RICH,

*11 Park Place, New York 7, N. Y.*

The applicant's letter of abandonment of March 21, 1951, has been received. Accordingly this application is held to be abandoned and will be forwarded to the abandoned files.

WILLIAM G. WILES, *Examiner.*

In the United States Patent Office

IN RE THE APPLICATION OF FREDERICK C. BERSWORTH

Filed July 17, 1948. Serial No. 39,377. For: Hydroxy Derivatives of Alkylene Mono- and Poly-Amines and Method of Forming Same.

405 LEXINGTON AVENUE,

*New York 17, N. Y.,*

*January 24, 1956.*

HON. COMMISSIONER OF PATENTS,

*Washington 25, D. C.*

SIR: Please allow Darby & Darby, Registration No. 10954, access to the abandoned file of the application above identified for the purpose of making copies of such parts thereof as they may specify.

Respectfully,

FREDERICK C. BERSWORTH,

*Applicant.*

39377

PAPER No. ——
JUL 27 1948
DIVISION 8

# 1948

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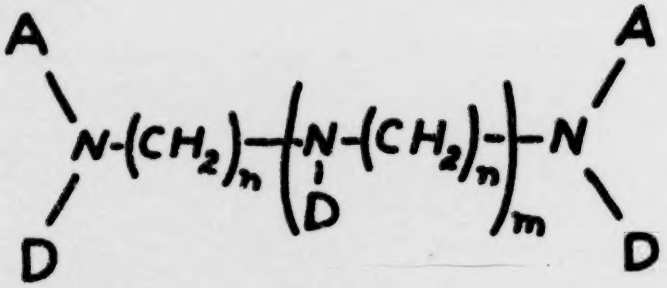
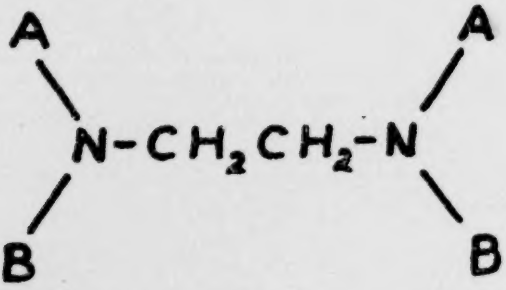
*Plaintiffs' Exhibit No. 8, C. A. 1637–54*

**CASE–B**
S.N. 39,377



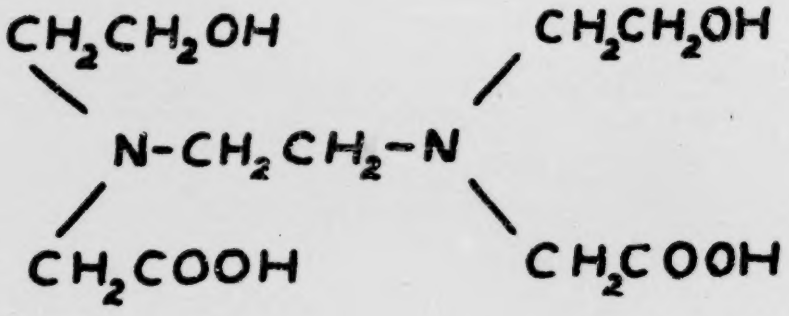

*Plaintiffs' Exhibit No. 9, C. A. 1637–54*

**CASE–B**
S.N. 39,377




*Plaintiffs' Exhibit No. 10, C. A. 1637–54*

**CASE–B**
S.N. 39,377



*Plaintiffs' Exhibit No. 12*

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE

Filed September 18, 1956. Harry M. Hull, Clerk.

*To all persons to whom these presents shall come, Greeting:*

This is to certify that the annexed is a true copy from the records of this office of the File Wrapper and Contents of the Application of Frederick C. Bersworth, filed, March 13, 1951, Serial Number 215,380, for Hydroxy Derivatives of Alkylene Polyamines.

In testimony whereof I have hereunto set my hand and caused the seal of the Patent Office to be affixed at the City of Washington, this eighteenth day of January, in the year of our Lord one thousand nine hundred and fifty-six and of the Independence of the United States of America the one hundred and eightieth.

ROBERT C. WATSON,
*Commissioner of Patents.*

Attest:

M. K. HANSEN,
*Certifying Officer.*

APPEAL NO. 33558

NUMBER (Series of 1948) 215380

PATENT NO.

DATED

DIV. 6

(EX'R'S BOOK)

Name FREDERICK C. BENSWORTH
Asso to The Dow Chemical Company, Midland Mich., a corp of Delaware.

of VERONA
State of NEW JERSEY
Invention HYDROXY DERIVATIVES OF ALKYLENE POLYAMINES

TITLE APPROVED

ORIGINAL

APPLICATION FILED COMPLETE MAR 13 1951

153261 PARTS OF APPLICATION FILED (Petition, Specification, Oath, First Fee $30) MAR 13 1951

Examined and passed for Issue , 19 Exr. Div.

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Final Fee , 19

Attorney MALCOLM N. NICH 11 PARK PLACE NEW YORK 7 N. Y.
~~Lucky & Nicholson 100 Lexington Ave. New York 16, N. Y.~~
Charles B. Watson Reg. #11,423 Lexington Ave. New York 16, N. Y.

Associate Attorney

No. of Claims Allowed Print Claim in O. G. Class

Division of App., No. , filed , 19

MALCOLM N. RICH
PATENT ATTORNEY
11 PARK PLACE, NEW YORK 7, N. Y.,
*Barclay 7-6066, March 12, 1951.*

Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

Re: Appln. of Frederick C. Bersworth, "Hydroxy Derivatives of Alkylene Polyamines."

SIR: I send herewith for filing an application for patent in this name. The enclosures are:

1. Petition, Specification, 4 Claims and Oath;
2. Check for $30.00 Filing Fee.

Respectfully,

MALCOLM N. RICH.

MNR:W.
Encls.

## Application for United States Letters Patent

### PETITION WITH POWER OF ATTORNEY

Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: Your Petitioner, Frederick C. Bersworth, a Citizen of the United States, and a resident of Verona, County of Essex, and State of New Jersey, and whose Post Office address is 46 Marion Road, Verona, New Jersey; prays that Letters Patent of the United States may be granted to him for improvements in hydroxy derivatives of alkylene polyamines set forth in the annexed specification.

And he hereby appoints Malcolm N. Rich, of 11 Park Place, New York City 7, New York (Reg. No. 13,142), his attorney with full power of substitution and revocation, to prosecute this application, to make alterations and amendments therein, to sign the draw-

ings, to receive the Letters Patent when issued, and to transact all business in the Patent Office connected therewith.

Signed at New York, N. Y., this 12th day of March, 1951.

Frederick C. Bersworth.
FREDERICK C. BERSWORTH.

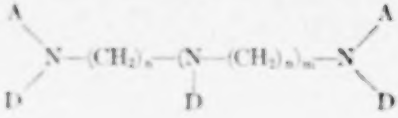
SPECIFICATION

*To All Whom It May Concern:*

Be it known that I, Frederick C. Bersworth, a Citizen of the United States, and a resident of Verona, County of Essex, and State of New Jersey, have invented certain new and useful Improvements in Hydroxy Derivatives of Alkylene Polyamines of which the following is a specification.

This invention relates to derivatives of alkylene polyamines and more particularly to the mono- and poly-hydroxy derivatives of said alkylene polyamines which derivatives are of wide utility in the art of chemistry and of chemical processes wherein compounds having strong detergent and wetting properties which are compatible with water soluble soap compounds and which in aqueous solutions form highly stable complexes with metal ions present in the solution, are desirably employed.

This application is a continuation-in-part application of my application Serial No. 39,377 filed July 17, 1948, which was a continuation-in-part of application Serial No. 657,893, filed March 28, 1946, in which first filed application was disclosed and claimed alkylene polyamine derivatives corresponding to the following structural formula:



wherein n represents one of the numerals 2 and 3; m has a value selected from the group consisting of zero (0), 1, and any multiple of 1; D is one of the substitute groups consisting of $-CH_2COOH$, $-CH_2CH_2COOH$, and a basic metal salt thereof; and wherein A is a hydrocarbon group containing at least two (2) carbon atoms and having not more than two (2) hydrogen atoms thereof replaced by one of the substituent groups consisting of $-CH_2COOR$, $-(CH_2)_2COOR$, $-CH_2COOM$, $-SO_3H$, $-SO_3M$, $-OR$, and $-OR'OR$ wherein R is alkyl or hydrogen, M is an alkali metal, and R' is alkylene, said groups being spaced at least two carbon atoms from the nitrogen atoms.

The present invention relates to compounds conforming to this general formula in which A consists of a mono- or poly-hydroxy hydrocarbon group, the compound otherwise conforming to the formula above given.

I have discovered that these mono- and poly-hydroxy derivatives of alkylene polyamines are highly effective sequestering agents in aqueous solutions for metal ions present therein and are particularly effective in sequestering polyvalent metal ions having a valency of at least three (3) forming complexes therewith which resist decomposition and precipitation of the metal ions by normal precipitating agents therefor over a wide range of temperatures and over a wide range of pH; and that said complexes are compatible and non-reactive with water-soluble soap compounds present in the solution.

The acidic hydroxy derivative compounds of the present invention are best formed by carboxylating a mono- or poly-hydroxy polyamine in accordance with the method described and claimed in my prior patent No. 2,407,645 issued September 17, 1946, acidifying

the reaction solution to neutralize the alkali metal salt thereof and to precipitate the hydroxy-amino acid therefrom, concentrating the solution to small volume to recover additional amounts of the acid, and separating the precipitated amino acid product from the solution. Alternatively, the alkali metal salt of the hydroxy-amino acid present in the reaction solution may be recovered therefrom by evaporating off the major portion of the water of hydration to form a concentrated solution of the salt and dehydrating the salt preferably under vacuum. This salt upon resolution in water and on neutralization with a strong mineral acid precipitates the hydroxy-acid as before described.

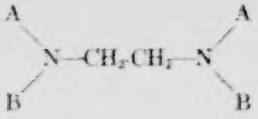
Mono- and poly-ethanol polyamines containing at least one *displaceable amino* hydrogen, but preferably more than one said amino hydrogen, are generally subject to carboxymethylation substitution in accordance with the process of my above identified patent to form mono- and poly-carboxylic (acetic) acid salts analogous to those identified above.

Similar and substantially equivalent chemical compounds are obtained on carboxymethylation of mono- and poly-propanol polyamines.

For the purposes of the present invention, however, I have found that while all of these ethanol and propanol derivatives of polyamines are chemically equivalent compound the effectiveness and efficiency of those compounds as complex-forming agents toward metal ions in solution therewith varies with variation in composition.

Of all of these compounds I have found that the most stable and reliable compounds for general purpose use especially in the presence of water-soluble soap compounds are those derivatives of ethylene di-

amine wherein two (2) of the amino hydrogens of the diamine have been displaced by ethanol or propanol and the other two (2) of the amino hydrogens have been displaced by acetic acid groups. Such a compound is indicated by the following formula:




wherein A represents ethanol ($-CH_2 \cdot CH_2 \cdot OH$) or propanol ($-CH_2 \cdot CH_2 \cdot CH_2 \cdot OH$) and B represents an acetic acid group ($-CH_2 \cdot COOH$) wherein the H may be displaced by substantially any basic metal ion but preferably is displaced by a strongly basic metal ion such as an alkali or alkaline earth metal ion.

This compound is formed by carboxymethylating di-ethanol or di-propanol ethylene diamine in accordance with the process of my prior patent, above identified, to displace the remaining two (2) amino hydrogens of the diamine with acetic acid groups ($-CH_2 \cdot COONa$). Where the process is properly conducted the reaction is substantially quantitative where the materials entering into the process are substantially pure. On a commercial basis yields approximating 90% to 95% of the theoretical may be consistently obtained.

One mole (148 g.) of redistilled diethanol ethylene diamine is dissolved in an equal volume of water and 5 g. of caustic soda are added. The resulting nearly colorless solution is heated in a boiling water bath and stirred vigorously during the slow (4-hour) addition of a saturated aqueous solution of 100 g. of sodium cyanide and 205 g. of a 30% formaldehyde solution. The addition of the cyanide is begun immediately and the addition of the formaldehyde is begun 15 minutes later, the proportional rate of addition

being the same so that the cyanide is completely added 15 minutes before the formaldehyde. Almost immediately after the addition of the first formaldehyde ammonia is given off and is vented through a reflux condenser at atmospheric pressure. For two hours after the completion of the formaldehyde addition the reaction mixture (now a pale yellow) is refluxed gently with vigorous stirring while a stream of air is passed slowly through it.

In commercial practice I prefer to first recover the alkali metal salt of this di-ethanol (or di-propanol) ethylene diamine diacetic acid from the reaction solution by evaporating off the major portion of the water of solution until the solution has a syrupy consistency and careful drying or dehydrating under vacuum.

The purified salt is then re-dissolved in water to form a 20%–30% solution and the anhydride thereof is obtained by acidifying the solution with a strong mineral acid ($H_2SO_4$ or HCl) to a pH of about 6.5 at which the anhydride is the least soluble.

As an example of the effectiveness and utility of the di-ethanol ethylene diamine diacetic acid compound as a sequestering agent for polyvalent metal ions in aqueous solutions therewith, the following test results are given:

As is well known in the art most commercial waters contain at least a small amount of iron. This iron will precipitate out of solution when the pH of the solution is made alkaline. In the many chemical processes conducted in alkaline pH solutions it is often desired to inhibit and prevent the precipitation of such basic iron compounds. Typical of these chemical processes is that involving the washing or laundering of organic fibrous materials wherein a water-

soluble soap, such as an alkali metal-fatty acid soap compound is employed. In such operation the iron content of commercial water is generally precipitated either in the form of ferric hydroxide or as an insoluble iron-fatty acid soap compound which compounds usually become affixed to the surface of the washed or laundered material requiring special acid treatment to be removed therefrom.

In the case of one sample of commercial water containing, by chemical analysis, 4 parts per million of Fe $^{+++}$, the addition of sodium hydroxide in an amount providing a 3% sodium hydroxide solution with heating to the boiling point was sufficient to precipitate the iron content of the solution as ferric hydroxide ($Fe(OH)_3$). The addition of a small amount of an alkali metal-fatty acid soap to the solution, in substitution for the sodium hydroxide, with heating was sufficient to form a precipitate of the insoluble iron-fatty acid soap.

By the addition, to the commercial water, of an amount of di-ethanol ethylene diamine diacetic acid providing approximately one molar weight of the acid per molar weight of the iron, calculated as Fe, the precipitation of $Fe(OH)_3$ and the formation of insoluble iron-fatty acid soap compounds is completely inhibited, even in the presence of sufficient free caustic alkali to increase the pH to as high as 12 and on extended heating at the boiling point of the solution or on several days standing.

Similar test results were obtained in water containing various other metal ions commonly found in commercial waters and the complexes formed by all of these metal ions with di-ethanol ethylene diamine diacetic acid or the alkali metal salts thereof, appear completely stabilized against precipitation by the

usual precipitating agents therefor over a wide range of pH and over a wide range of relative concentrations of precipitant to complex.

The exact composition of the complex formed with these metal ions by di-ethanol ethylene diamine di-acetic acid is the subject of conjecture but it is believed that these metal ions must be held in un-ionized complex combination by the residual valency bonds of the amino nitrogens set free and made active upon solution and ionization of the ethanol amino acid in water.

Where the hydroxy-poly-amino acid contains but one ethanol substituent group and three acetic acid substituent groups, such as is present in mono-ethanol ethylene diamine tri-acetic acid, the effectiveness of the compound towards some polyvalent metal ions is somewhat different. In the case of iron, for example, the complex formed appears to be primarily one holding the iron in its ferric state whereas the di-ethanol amino acid appears to hold the iron in its ferrous state.

To a 50% aqueous solution of 104 g. monoethanol ethylene diamine is added 5.0 g. of caustic soda. The resulting solution is heated between 95°–100° C. and vigorously stirred for five hours, while 150 g. of sodium cyanide and 92 g. of formaldehyde are slowly added. The sodium cyanide is added in the form of a nearly saturated solution while the formaldehyde is added as a 30% aqueous solution, care being taken never to allow the stoicheiometric amount of cyanide added to be exceeded by the formaldehyde. The reaction is "finished off" by heating and stirring at slow reflux for two hours, and passing through a slow stream of air to assist in the removal of ammonia. If the elimination of ammonia is not complete at the

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end of this time, it is advisable to distill off a small amount of the solvent.

The reaction product may be isolated as a salt by method (1) or as an acid lactone by method (2):

(1) The reaction mixture is evaporated to half its volume (or to a viscous liquid) under partial vacuum and then extracted with ethanol (to remove excess caustic alkali). Further careful evaporation results in the isolation of hydrated trisodium salt of monoethanol ethylene diamine triacetic acid in better than 90% purity. The chief contaminant is sodium carbonate (along with a smaller amount of the trisodium salt of ammonia triacetic acid), which is of no disadvantage for most uses of the product. It may be largely be avoided, however, by preventing the access of carbon dioxide to the reaction mixture by suitable absorbents.

(2) The pH of the reaction mixture is reduced to about 2.0 with sulfuric acid, whereupon the lactone of monoethanolethylene diamine triacetic acid is obtained as a crystalline precipitate on evaporation of the solution. Although contaminated with sodium sulfate, it may be purified by further fractional crystallization.

Di-propanol ethylene diamine di-acetic acid and mono-propanol ethylene diamine tri-acetic acid react substantially identically to that above described in connection with the di-ethanol and mono-ethanol amino acids.

Similarly, other mono- and poly-ethanol and propanol poly-alkylene polyamines form ethanol and propanol acetic acid derivatives which are substantial equivalents to those hereinabove described, insofar as their complex-forming properties with metal ions in aqueous solutions may be concerned, and by the term ethylene diamine as it hereinafter appears in the

claims these other polyamines are meant to be included.

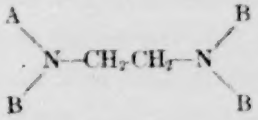
One of the major advantages of the complex-forming compounds of the present invention, over those heretofore employed in the art, is that the compounds themselves and the metal complexes thereof are soluble in water over a wide range of pH, from strongly acid to strongly alkaline, due to the presence therein of the non-ionic solubilizing ethanol and propanol groups, and due to the fact that the weakly ionic carboxylic groups upon complex-formation become substantially non-ionic.

Many of these mono and poly hydroxy polyamino acids may be recovered from concentrated aqueous solutions as anhydrides upon acidification with a strong mineral acid to a pH at which dehydration of the amino acid occurs. Other means for dehydrating the amino acid may be employed to recover same from aqueous solution. However, in the usual small amounts of these amino acids required to sequester the metal ions present in most commercial waters the presence of relatively large amounts of free acid or free alkali in the solution is ineffective in causing decomposition of the hydroxy-amino acids, their salts or the metal complexes thereof.

Having hereinabove generically and specifically disclosed the present invention and having given several specific examples of the same and indicated the extent of utility thereof in one field of use, it is believed apparent that the invention may be widely modified without essential departure therefrom and all such modifications and departures are contemplated as may fall within the scope of the following claims:

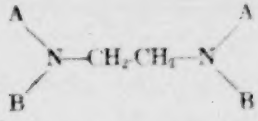
What I claim is:

1. The compound conforming to the formula—




wherein A is a substituent group selected from the group consisting of ethanol and propanol and B is an acetic acid substituent group.

2. The compound conforming to the formula—




wherein A is a substituent group selected from the group consisting of ethanol and propanol and B is an acetic acid substituent group.

3. The basic metal salts of the compound of claim 1.

4. The basic metal salts of the compound of claim 2.

In witness whereof, I have hereunto signed my name.

Frederick C. Bersworth.

FREDERICK C. BERSWORTH.

OATH

STATE OF NEW YORK,

*County of New York, ss:*

Frederick C. Bersworth, the above-named Petitioner, being duly sworn, deposes and says that he is a Citizen of the United States, and a resident of Verona, County of Essex, and State of New Jersey; that he verily believes himself to be the original, first and sole inventor of the Improvements in Hydroxy Derivatives of Alkylene Polyamines described and claimed in the annexed specification; that he does not know and does

not believe that the same was ever known or used before his invention or discovery thereof, or patented or described in any printed publication in any country before his invention or discovery thereof, or more than one year prior to this application; that, as to the portions of his invention that are common to this application and of his prior filed application Serial No. 39,377 filed July 17, 1948, which application was a continuation-in-part of application Serial No. 657,893 filed March 28, 1946, now issued as U. S. Patent No. 2,524,218 dated October 3, 1950; he does not know and does not believe that the same were in public use or on sale in the United States for more than one year before the respective filing dates of said prior filed applications; that, as to the portions of this invention that are described and claimed in said prior files applications Serial No. 39,377 and Serial No. 657,893 (now Patent No. 2,524,218) he does not know and does not believe that the same were in public use or on sale in the United States for more than one year prior to this application; that said invention has not been patented in any country foreign to the United States on an application filed by him or his legal representatives or assigns more than twelve months prior to this application; and that no application for patent on said improvements has been filed by him or his legal representatives or assigns in any country foreign to the United States.

Frederick C. Bersworth.

FREDERICK C. BERSWORTH.

Sworn to and subscribed before me this 12th day of March, 1951.

MURRAY COHN,
*Notary Public.*

Notary Public, State of New York, No. 24-5745400. Qualified in Kings County. Certified with New York

& Kings County Clerk's & Registers. Term expires March 30, 1952.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

POL–90

PAPER NO. 2

Address only the Commissioner of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the EXAMINER in charge of this application. John A. Marzall, Commissioner of Patents.

Division: 6—Room 7725. Applicant: Frederick C. Bersworth. Ser. No. 215,380. Filed: March 13, 1951.

For: Hydroxy Derivatives of Alkylene Polyamines.

MALCOLM N. RICH,
*11 Park Place, New York 7, N. Y.*

References made of record.

Ulrich et al.—1,973,860, Sept. 18, 1934, 260–534
Hoffman et al.—2,017,537, Oct. 15, 1935, 260–534.
Reppe et al.—2,195,974, Apr. 2, 1940, 260–534.
Bersworth—2,413,856, Jan. 7, 1947, 260–534.
Bersworth—2,428,353, Oct. 7, 1947, 260–534.
I. G. Farben (Ger.)—638,071, Nov. 9, 1936, 260–534.
I. G. Farben (Ger.)—642,244, Mar. 6, 1937, 260–534.

Claims 1, 2, 3 and 4 are rejected as being fully met by either of the Bersworth patents cited above which disclose the compounds of the claims. Applicant's patent No. 2,524,218 has been carefully considered but has not been found to have a disclosure sufficient to support the instant claims. Hence, the subject matter is held to be dedicated (Gladding McBean v. Clarke.

16 F (2nd) 50). There is a distinction between the disclosure required as a reference and that required as support for claims.

Claims 1 to 4 inclusive are rejected as being unpatentable over the German patent No. 642.244 or this patent in view of the Ulrich et al., Hoffman et al., or Reppe et al. patents, all cited above. The basic reference clearly discloses the class of materials and gives an example of a closely related compound (Ex. 6) indicating that applicant's compounds are of the class contemplated. Applicant has covered the remaining two possibilities. The other patents clearly set forth the class of materials. The compound of Ex. 6 of the German patent is as closely related to the compound of claim 2 as the compound of claim 1 is related to that of claim 2.

There being no generic claim, election is required as follows:

(I) To a tricarboxylic acid (1).

(II) To a dicarboxylic acid (2).

(III) To a salt of claim 1 (3).

(IV) To a salt of claim 2 (4).

Applicant is advised that his response to be complete under Rules 143 and 111 must include, in addition to a response to the rejection, an identification of the disclosed species that he elects consonant with the requirement, and a listing of all claims restricted or generic thereto. An argument that the generic claims are allowable, or that all claims are generic or amended to be generic, unless accompanied by an election, is non-responsive.

WILLIAM G. WILES,
*Act. Examiner.*

In the United States Patent Office

Applicant: Frederick C. Bersworth. Division: No. 6. Room: No. 7725. Serial No. 215,380. Filed: March 13, 1951. Invention: Hydroxy Derivatives of Alkylene Polyamines.

*Amendent (1)*

Hon. Commissioner of Patents,
*Washington, 25, D. C.*

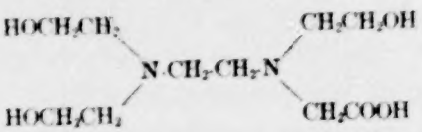
Sir: In response to the Official Action of July 2, 1951, reconsideration of the rejections made is respectfully requested in the light of the following traversing arguments:

(1) In the rejection of paragraph 1 of the Action, the Examiner proceeds on an erroneous major premise. This application is clearly identified on page 1 as being a continuation-in-part of application Serial No. 657,893 filed March 28, 1946 (now U. S. Patent No. 2,524,218) and the *disclosure* that follows and continuing to line 11 on page 2 is copied *verbatim* from the disclosure of this patent (page 1, column 1, lines 25 to 46 inclusive). The compounds herein claimed are those conforming to this generic formula wherein the *A groups* are the mono- or poly-hydroxy hydrocarbon groups.

In view of this fact, the rejection of paragraph 1 is clearly improper as based on a wrong major premise. The *genus disclosure* appears in the parent application. That is all that is required to form the basis for the instant application. Accordingly applicant's patents Nos. 2,413,856 and 2,428,353 are not prior art to the instant application.

Referring now to the rejection of claims 1 to 4 on art (paragraph 2 of Action) the attention of the

Examiner is directed to the fact that the German Patent 642,244 discloses triethanol ethylene diamine *monoacetic* acid as a gas absorbent:




which is the most closely related of the compounds listed to the compounds of the present application, but differs greatly from them both in chemical behavior and in chemical properties. All the remaining compounds listed in the German Patent 642,244 have secondary or primary amino groups and are not effective chelating agents of the type disclosed in the present invention.

All of the compounds disclosed in the Ulrich, et al., patent are amides (none of the compounds in the present application are amides) and are widely different in chemical properties. They are not, for example, effective sequestering or complexing agents. Further, none of the compounds from which these amides are prepared are sequestering agents. They are all monoamines and do not suggest the compounds of the present invention which are exclusively diamines.

The Hoffman, et al., compounds are not related to the compounds of the present application since:

(1) none of the compounds have more than one basic amino group (two in the present application);

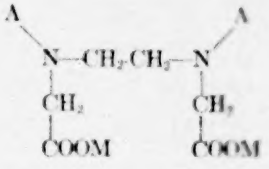
(2) all of the compounds have the basic amino group in the Beta position to the carboxyl.

The differences indicated above render the Hoffman compounds much weaker as complexing agents and deactivating agents for metal ions. In fact they are not satisfactory for the purposes described in the present application.

All of the Reppe compounds (except one) have basic amino groups attached to the carbon atom which is *beta* to the carboxyl group, and hence are subject to the same objections as the Hoffman compounds. The exception, aniline acetic acid, is not related to the compounds of the present invention.

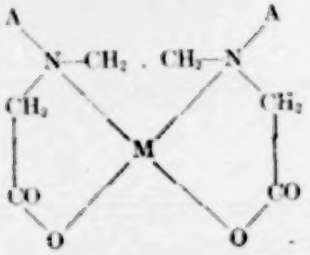
Referring to the last sentence of the 1st page and top of 2nd page of the Action: The Examiner's argument is traversed by the fact that the generic formula, page 1 of application, does not include the compound of the German patent, but does cover all the compounds claimed in the present invention. The differences between Example 6 of the German patent and the class of complexing agents claimed in the present application is best understood from the following considerations:

(1) According to the generic formula on page 1, the basic chelating structure essential in the compounds claimed for performing the functions described is:




wherein A represents ethanol or propanol; and M represents hydrogen or alkali metal; and the essential groups for performing the work are the two carboxyls with two *alpha* (not beta) basic nitrogens which are spaced by two or at the most three carbon atoms. When these essential groups are present (two acidic

and two basic), a chelate structure of the following type forms with metal ions:




This provides high stability to the structure, and cannot be formed by the compound of the German patent. The A groups provide solubility and auxiliary activity specific, or seemingly so, for the trivalent metal ions when hydroxyl groups are present.

In view of these clearly pointed out differences between the compounds of the instant invention and those of the references, the instantly claimed compounds are believed clearly patentable.

Pending the allowance of generic claims applicant elects to prosecute claims to the species dicarboxylic acid of claim 2.

A favorable action on reconsideration is respectfully solicited.

Respectfully submitted.

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His Attorney.*

DECEMBER 28, 1951.
MNR: W.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

POL–90

Address only the Commissioner of Patents, Washington 25, D. C.

PAPER NO. 4

All communications respecting this application should give the serial number, date of filing and name of the applicant.

Please find below a communication from the EXAMINER in charge of this application, John A. Marzall, Commissioner of Patents.

Applicant: Frederick S. Bersworth. Ser. No. 215,380. Filed March 13, 1951. For Hydroxy Derivatives of Alkylene Polyamines.

MALCOLM N. RICH,
*11 Park Place, New York 7, N. Y.*

Responsive to letter filed Dec. 29, 1951.

Claims 1, 2, 3 and 4 are again rejected as being fully met by either of the Bersworth patents of record which disclose the class of compounds of the claims. Applicant's patent No. 2,524,218 has been carefully considered but has not been found to have a disclosure sufficient to support the instant claims. To carry the date the disclosure must be sufficient to support claims to the subject matter. If the disclosure of the patent No. 2,524,218 were sufficient to support the claims, then by the same reasoning the German patent 642,244 anticipates the claims, since this patent has a generic disclosure of the subject matter. Accordingly, the Bersworth patents, which disclose the compounds, are a bar to the instant claims. There is a difference between the disclosure required as a reference and

that required as support for claims (*In re Stoll et al.*, 603 O. G. 378).

Claims 1 to 4 inclusive are again rejected as being unpatentable over the German patent No. 642,244 of record or this in view of *Ulrich et al, Hoffman et al* or *Reppe et al* patents of record. The basic reference clearly discloses the class of materials and gives an example of a closely related compound (Ex. 6) indicating that applicant's compounds are of the class contemplated. Applicant has covered the remaining two possibilities. The other patents set forth the class of compounds and related analogs. The compound of Ex. 6 of the German patent is as closely related to the compound of claim 2 as that of claim 2 is to that of claim 1. The salts appear to be anticipated. However, simple salts of acids are not patentably significant over the acids (In re Williams, 649 O. G. 912).

Claims 1, 3 and 4 are rejected as failing to read on the elected species.

This action is made *Final*.

WILLIAM G. WEIS,

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*Acting Examiner.*

In the United States Patent Office

Applicant: Frederick C. Bersworth. Division: No. 6. Serial No. 215,380. Filed: Mar. 13, 1951. Invention: Hydroxy derivatives of alkylene polyamines.

## Reconsideration Request

Hon. Commissioner of Patents,
*Washington 25, D. C.*

Sir: In response to the Official Action of January 22, 1952, the Examiner is respectfully requested to reconsider the Final Rejection of record and to withdraw same as being premature and as being based primarily upon an error of law which is clearly reversible on appeal.

It is urged that the Examiner in the rejection of paragraph 1 of the Action has failed to recognize the continuity of subject matter presented by this application, its predecessor application Ser. No. 39,377 filed on *July 17, 1948* (now abandoned) which was a continuation-in-part of the parent application Ser. No. 657,893 filed *March 28, 1946*, which issued as Patent No. 2,524,218.

On the basis of this *continuity* of subject matter, applicant is clearly entitled to the benefit of the filing date of March 26, 1946, for priority, thereby effectively antedating all prior patents cited having an issue date that is not more than one (1) year prior to this priority date.

The attention of the Examiner is directed to the fact that the *generic* compound disclosed in the parent patent includes the *species* compounds now claimed in this application. This is sufficient to give the applicant the benefit of the filing date of the parent patent in this application.

The Examiner is misled by the fact that the filing date of this application is subsequent to the issue date of the parent patent. The fact that the immediate parent application of this application was copending at the date of issue of the parent patent negatives any

presumption of abandonment of all subject matter contained in this copending application.

Referring to the *disclosure* of the now abandoned predecessor application Serial No. 39,377 the attention of the Examiner is directed to the fact that it was directed to cover those *species* derivatives of the alkylene polyamino acid compounds *generically* disclosed in the parent patent 2,524,218 and identified by the genus formula in column 1 on page 1 of the patent containing the —OR'OR groups, and particularly to those compounds of the generic formula wherein the A groups consist of mono or poly hydroxy hydrocarbon groups. (See page 2, paragraph 2, lines 11–14.)

The *specific examples* disclosed in this application Ser. No. 39,377 (page 3) is the ethanolamine di-carboxylic acid and (page 4, paragraph 1) is the mono ethanol ethylene diamine triacetic acid and the di-ethanol ethylene diamine diacetic acid. On page 4 at line 17 is mentioned the ethanol and *propanol* derivatives of mono and poly amines. At the top of page 5 the specific formula of rejected claim 2 is fully disclosed. Claim 5 of this application is moreover directed to cover the compound of rejected claim 2.

In the face of this disclosure and clear effort by applicant to claim the compounds herein claimed appearing in this predecessor application which was copending with the parent patent at the date of issue of the parent patent, the *Examiner's position* with respect to this parent patent being a bar to the allowance of claims to these compounds in this application *is clearly untenable*, and this ground for final rejection accordingly should be withdrawn.

It is urged that the Examiner, in view of the failure to consider the disclosure of application Serial No. 39,377 before making final the rejection of paragraph

1 of the Office Action has failed to give sufficient consideration to the remarks of applicant traversing the rejection on art made in the previous amendment (1). Otherwise, the rejection of the first Action would not have been repeated *verbatim* in the second Action.

Accordingly, it is respectfully requested that the final rejection of record be withdrawn and that the Examiner should re-open prosecution of the application giving applicant a second Action on art based on the traverse presented in previous amendment (1).

It is urged that in view of the traversing argument claims 1 to 4 are fully and clearly allowable to applicant over the disclosures of these prior art references.

A favorable action on reconsideration and prompt allowance of the rejected claims is respectfully solicited.

Respectfully submitted.

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His Attorney*

JUNE 6, 1952.
MNR: W.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

PAPER NO. 6

POL–90

Address only the Commissioner of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the EXAMINER in charge of this application. John A. Marzall, Commissioner of Patents.

Applicant: Frederick C. Bersworth. Ser. No. 215,380. Filed March 13, 1951. For Hydroxy Derivatives of Alkylene Polyamines.

MALCOLM N. RICH,
*11 Park Place, New York 7, New York.*

Responsive to amendment filed June 7, 1952.

The applicant's letter of June 7, 1952 has been received and carefully considered but has not been found to overcome the rejections of the claims. The date of Serial Number 39,377 does not overcome the dates of the Bersworth patent and Serial Number 657,893 (patent No. 2524218) is not considered to have adequate support for the claims, as of record.

*The six months period for response to the final rejection runs from January 22, 1952.*

H. SURLE,
*Examiner.*

In the United States Patent Office
(Div. 6)

RE: APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 215,380. Filed: March 13, 1951. For: Hydroxy Derivatives of Alkylene Polyamines.

The COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: Applicant hereby appeals to the Board of Appeals from the decision of the principal examiner finally rejecting claims 1, 2, 3, and 4.

Respectfully submitted.

Malcolm N. Rich.
MALCOLM N. RICH,
*Attorney for Applicant.*

JULY 21, 1952.

405368—56——17

## In the United States Patent Office

## Before the Board of Appeals, Appeal No. 33,558

### In re Application of Frederick C. Bersworth

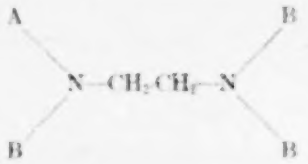
Serial No. 215,380. Filed: March 13, 1951. For: Hydroxy Derivatives of Alkylene Polyamines.

### Brief for Bersworth

This is an appeal from the action of the Examiner in finally rejecting all of the claims of this application for the reasons of record and on the art of record, the Examiner being, it is believed, in clear error in so rejecting the claims.

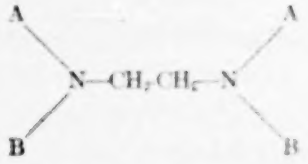
### THE REJECTED CLAIMS

1. The compound conforming to the formula—




wherein A is a substituent group selected from the group consisting of ethanol and propanol and B is an acetic acid substituent group.

2. The compound conforming to the formula—




wherein A is a substituent group selected from the group consisting of ethanol and propanol and B is an acetic acid substituent group.

3. The basic metal salts of the compound of claim 1.

4. The basic metal salts of the compound of claim 2.

THE ART OF RECORD

Ulrich et al.—1,973,860, Sept. 18, 1934, 260–534.
Hoffman et al.—2,017,537, Oct. 15, 1935, 260–534.
Reppe et al.—2,195,974, Apr. 2, 1940, 260–534.
Bersworth—2,413,856, Jan. 7, 1947, 260–534.
Bersworth—2,428,353, Oct. 7, 1947, 260–534.
I. G. Farben (Ger.)—638,071, Nov. 9, 1936, 260–534.
I. G. Farben (Ger.)—642,244, Mar. 6, 1937, 260–534.

THE REJECTION

The Final Rejection of record states two (2) grounds for rejecting the appealed claims:—

(1) As being fully met by either of the two (2) Bersworth patents of record.

(2) As being unpatentable over the German patent No. 642,244 or over this patent in view of Ulrich et al., Hoffman et al., or Reppe et al.

Claims 1, 3 and 4 are additionally rejected as not readable on the elected species.

In explanation of the first ground for rejection the Examiner holds that the *disclosure* of the parent application to this application (now U. S. patent No. 2,524,218) is insufficient to support the rejected claims and that applicant, accordingly, is not entitled to the filing date of this parent application for priority over these two patents.

In explanation of the second ground for rejection the Examiner holds that the basic reference clearly discloses the *class* of materials and gives an example of a closely related compound (Ex. 6) indicating that applicant's compounds are of the class contemplated, and that the other patents set forth the class of compounds and related analogs.

ARGUMENT

The Examiner's first ground for rejection raises a question of law and the second ground raises a question of fact.

The question of law presented in the first ground for rejection is whether or not an applicant who makes a genus disclosure in a parent application is entitled to the benefit of the filing date of the parent application for priority as to species disclosed and claimed in subsequently filed continuation applications. *The answer to this question is believed clearly in the affirmative.*

U. S. patent No. 2,524,218, which issued on October 3, 1950, has a filing date of March 28, 1946.

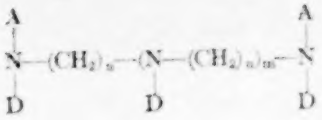
The present application was filed on March 13, 1951, as a continuation-in-part of application Serial No. 39,377, filed on July 17, 1948 (now abandoned).

Application Serial No. 39,377 was filed as a continuation-in-part of the parent application now issued as U. S. patent No. 2,524,218.

Applicant in rejected claims 1 and 2 of this application claims the *mono- and di-ethanol and propanol* ethylene diamine tri- and di-acetic acids.

In the now abandoned parent application Serial No. 39,377 applicant attempted to claim genus and species mono- and poly-hydroxy alkylene polyamine poly-acetic acids, of which mono- and di-ethanol and propanol ethylene diamine tri- and di-acetic acids are species described and claimed therein.

In the preceding parent application (Patent No. 2,524,218) the *generic* compound conforming to the formula was disclosed:—



wherein n is either 2 or 3 and M is either zero or a positive integer and wherein D is a member of the group consisting of $CH_2$-COOH, $CH_2$-$CH_2$-COOH, and their alkali metal salts and wherein A is a hydrocarbon group containing at least two carbon atoms and having not more than two hydrogen atoms replaced by a member of the group consisting of $CH_2$-COOR, $(CH_2)_2$-COOM, $SO_3H$, $SO_3M$, OR and OR'OR wherein R is one of the group consisting of alkyl and hydrogen, M is an alkali metal, R' is an alkylene group, said replacing groups being spacted from the N atoms of the compound by at least two carbon atoms of the hydrocarbon group containing the same.

In this genus disclosure the compound containing an A group conforming with the OR'OR species where R is hydrogen is clearly a hydroxy compound. From the *disclosure* of the first filed continuation-in-part application (Serial No. 39,377) it is clearly apparent that applicant, as of the date of filing of this first filed application (July 17, 1948) had every intention of reserving to himself this genus disclosure of the parent application in so far as it relates to mono or polyhydroxy hydrocarbon groups comprising the A groups of the genus. (See page 2, lines 11 to 24 inclusive of Serial No. 39,377.)

The rule is well-established in the U. S. patent system, and needs no citation of decisions to support it, that any subject matter disclosed but not claimed in an issued patent is presumed dedicated to the public *unless* the patentee in a continuation application filed prior to the issue of the patent has negatived this presumption of abandonment by continuing to claim this unclaimed subject matter.

Applicant, in conformity with this rule, has obviously negatived any presumption of abandonment to the mono and polyhydroxy compounds falling within

the genus disclosure of patent 2,524,218 by filing the continuation-in-part application Serial No. 39,377 prior to the issue of this U. S. patent. An examination of the original claims of this application Serial No. 39,377 discloses that applicant in claim 1 claims genus mono and polyhydroxy compounds conforming to the genus formula of patent 2,524,218; in claim 2 claims the species compounds derived from ethanol and propanol; and in claims 5 and 6 claims the species compounds di-ethanol and di-propanol ethylene diamine diacetic acid.

Accordingly, as of the date of filing of this first continuation-in-part application (July 17, 1948) genus and species hydroxy compounds falling within the genus disclosure of the parent application are being claimed by applicant as his invention.

The application on appeal was filed on March 13, 1951, as a continuation-in-part of application Serial No. 39,377 and further prosecution of this parent application was abandoned in favor of the present application. The claims of record in the instant application are limited to the ethanol and propanol ethylene diamine-polyacetic acid species disclosed and claimed in application Serial No. 39,377.

The fact that these claims were being made in the application 39,377 prior to the issue of patent 2,524,218 and were continuously asserted therein after the issue of this patent, and that these claims were carried forward into the instant application, clearly negatives any intention on the part of applicant to abandon this invention to the public on issue of patent 2,524,218. The Examiner, accordingly, is in clear error in rejecting the claims of record as stated under the first grounds for rejection.

By limiting the claims of this application to species applicant can only be presumed to have abandoned his

statutory right to a claim covering genus which he preserved by filing application Serial No. 39,377 prior to the issue of patent 2,524,218.

By disclosing and claiming the species now claimed in this application in his application 39,377 at a date prior to the issue of patent 2,524,218 applicant has effectively negatived any presumption of abandoment of this species on issue of this patent.

The fact that the instant application was filed at a date subsequent to the issue of patent 2,524,218 is wholly immaterial as applicant in application Serial No. 39,377 prior to the issue of this patent was asserting claims to this species and has continuously asserted these claims to date.

To answer the Examiner's rejection more specifically, the fact that applicant in patent 2,524,218 disclosed genus and that he in application Serial No. 39,377 disclosed the same genus and the species now claimed in the application on appeal completely rebuts any presumption of abandonment of this species on issue of patent 2,524,218.

It is submitted that he Examiner fails to recognize that there is a great difference between abandonment on issue of a patent and abandonment by voluntary act. Applicant has abandoned his effort to obtain an allowance of genus claims commensurate with the scope of the disclosure in application Serial No. 39,377 and has voluntarily limited his claims and his disclosure in this application to *species*.

This voluntary limitation of claims to species, however, does not operate to deprive applicant of the priority date of March 26, 19[illegible], the filing date of patent 2,524,218 to which he is entitled for both genus and species. This is obviously correct as genus *includes* all species whereas the reverse is not true.

As an illustration of this, in the situation where an applicant first discloses species and later files on genus, proof of reduction to practice of the species disclosed in the first filed applicatoin cannot be accepted as proof of reduction to practice of genus (In re Steenbock, C. C. P. A. 23, 1244). The evidence of record is against the presumption that genus and species are one and the same invention. Where genus is disclosed applicant is not compelled *under the Statute* to disclose more than one species to support genus and all other species are presumed to have been simultaneously disclosed or invented for purposes of priority.

This is the situation here involved and the Examiner's first ground for rejection is clearly in error and must be reversed as the *issue* dates of the two Bersworth patents relied on are each subsequent to the filing date of applicant's parent patent 2,524,218. Such action is respectfully solicited on this first ground for rejection.

REFERRING TO THE SECOND GROUND FOR REJECTION

The German patent 642,244 merely discloses *triethanol* ethylene diamine monoacetic acid as a gas absorbent. This compound is neither a homologue, analogue or a compound having analogous chemical properties to the compounds herein claimed.

The compounds of the instant invention are metal ion-sequestering agents. The compound disclosed in this German patent is *not* a sequestering agent. It is only when one of the amino hydrogens of *each* amino group in ethylene diamine has been displaced by acetic acid (or its equivalent acid group) that metal ion-sequestering properties appear in the compound. The German patent fails to disclose this fact and being a foreign reference must be limited for purposes of

anticipation to the precise disclosure appearing therein.

Considered alone, therefor, the compounds of the instant invention are not anticipated by this reference.

When the disclosure of this German reference is taken with Ulrich, Hoffman or Reppe, anticipation also is lacking. Contra to the Examiner, the German reference fails to disclose the *class* of materials as it fails to disclose a compound analagous to those herein claimed which is capable of reacting as a metal ion-sequestering agent. *Neither Ulrich, Hoffman or Reppe can be construed as teaching one skilled in the art to convert the compound of the German reference into a metal ion-sequestering agent.*

Ulrich, for example, merely disclosed amides which are completely different chemical compounds from those disclosed by the German patent or by applicant which amide compounds, per se, are not sequestering agents.

Hoffman discloses compounds totally unrelated to the instantly claimed compounds. None of the Hoffman compounds have more than one basic amino group and all of the Hoffman compounds have this basic amino group in beta position to the carboxyl. Wherein such compounds can be used to modify the compound of the German reference is totally unapparent and unexplained by the Examiner.

Substantially the same situation is true with the compounds of the Reppe patent. All of the compounds of this patent, with a single exception, show a basic amino group in beta position to the carboxyl group. The single exception is aniline acetic acid which is remote from the instantly claimed compounds. The Examiner fails to give any reasonable statement explaining *wherein* the compounds of the German patent can be modified by the teaching of

Reppe to be substantially identical to those claimed by applicant.

It is respectfully urged that while it is recognized that the burden of proof of patentability rests on applicant he should not be called upon to bear an unreasonable burden occasioned by the unreasonable citation of non-pertinent references by the Examiner. In chemical cases it is difficult enough to differentiate patentably from pertinent references.

In this instance, it is *totally unapparent* wherein the compound of the basic reference which, per se, is non-anticipating can be modified, changed or converted into an anticipating compound by the teaching of any one of the supplemental references relied on by the Examiner and the Examiner in his rejection fails utterly to explain how this can be done, in violation of the rules of practice. The Examiner is called on to do more than give "lip service" to the rules of practice in giving any rejection on art.

In view thereof, it is urged that in the absence of pertinent prior art the rejected claims are clearly allowable to applicant.

CONCLUSION

Having hereinabove shown that the Examiner's rejection on both grounds is in clear error, it is urged that applicant is entitled to an allowance of all claims on appeal and such action is respectfully solicited.

Respectfully submitted.

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His Attorney.*

SEPTEMBER 12, 1952.
MNR:R.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Address only: the Commissioner of Patents, Washington 25, D. C.

PAPER No. 9

Appeal No. 33,558 Before the Board of Appeals

All communications respecting this application should give the serial number, date of filing, name of the applicant, and appeal number.

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Ser. No. 215,380. Filed March 13, 1951, For Hydroxy Derivatives of Alkylene Polyamines.

Malcolm N. Rich for Appellant.

EXAMINER'S STATEMENT

This is an appeal from the final rejection of claims 1, 2, 3 and 4, all the claims in the application. Claims 1, 3 and 4 are rejected as failing to read on the elected subject matter.

A correct copy of the appealed claims appears on page 1 of the applicant's brief.

The references relied on have been listed in applicant's brief, page 2.

The subject matter is adequately described in the claims on appeal and below.

THE REFERENCES

The Bersworth patents disclose on page 1 of each the generic formulae covering the compounds claimed. The formula is considered to be definite enough to suggest the *class* of compounds claimed to a chemist

of average skill. The usual alkylene group for "Z" in the formulae is ethylene, since ethylene diamine is the most common of the diamines. The definition of y and y′ as "an alkylol" group is considered to be suggestive of the lowest members, methylol and ethanol (claimed as a *class*).

*The German Patent No. 642,244* discloses a generic class of materials and a specific compound under the class (page 7) N,N′-tri(hydroxy ethyl) ethylene diamine-N-acetic acid, indicating that this class of compounds is contemplated under the generic class described. Of the ethanol substituted members of the class, the claims at bar cover the other two possibilities.

THE REJECTIONS

The final rejection (paper No. 4) is considered to be sound, and is adhered to.

As to the critical difference in disclosure required to support a claim and that required as a reference, attention is urged for the decision In re Stoll et al. (603 O. G. 378). Attention is also called to In re Bersworth (650 O. G. 628).

The rejections are considered to be sound.

H. SURLE,
*Examiner.*

MALCOLM N. RICH,
*11 Park Place, New York 7, N. Y.*

In the United States Patent Office

Appeal No. 33,558 Before the Board of Appeals

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 215,380. Filed: March 13, 1951. For: Hydroxy Derivatives of Alkylene Polyamines.

## Petition To Remand

Your Petitioner, the applicant in the above application, hereby respectfully petitions the Board of Appeals to permit the entry of the attached affidavits by Frederick C. Bersworth and Arthur E. Martell in the record of this application and for a remand of the application to the Primary Examiner for reconsideration and revision of the Examiner's Statement in view of said affidavits or for an allowance of the appealed claims if such action is now warranted in view of said affidavits.

Your Petitioner avers that this petition and attached affidavits have not been filed for the purpose of delay but for the purpose of clarifying the appeal record on the two (2) grounds for final rejection of the appealed claims and to negative the new grounds for rejection raised in the last paragraph on page 2 of the Examiner's Statement.

As the Hearing date for this appeal has not been set, this Petition to Remand is believed proper under the provisions of Rule 195 and a favorable action thereon is respectfully requested.

Respectfully submitted.

Frederick C. Bersworth,
By Malcolm N. Rich,
*His Attorney.*

December 31, 1952.
MNR: W.
Encls.

In the United States Patent Office

Before the Board of Appeals

Appeal No. 33,558

Division 6

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 215,380. Filed: March 13, 1951. For: Hydroxy Derivatives of Alkylene Polyamines.

AFFIDAVIT

STATE OF NEW YORK,
*County of New York.*

Frederick C. Bersworth, being duly sworn, deposes and says:

That the subject matter incorporated in the Exhibit Affidavit accompanying this affidavit, and forming a part hereof, in a substantial measure, involves a discussion of basis chemistry of the type of chelating agent embodied in patent application Serial No. 215,380. The discussion of the chemistry of these materials is necessary in order to give accurate significance to the presentation of experimental subject matter appearing at pages 10, 11 and 12 of the said affidavit, which experimental subject matter was carried out and developed by Dr. Arthur E. Martell;

That the subject matter has not heretofore been incorporated in the record because it was not in existence in usable form and even at this stage it is only a part of a comprehensive program of funda-

mental study of chelating agents being carried out under his auspices.

FREDERICK C. BERSWORTH.

Subscribed and sworn to before me this 19th day of December 1952.

[SEAL] EILEEN LAMERSON,
*Notary Public.*

Eileen Lamerson, Notary Public, State of New York. No. 24–7410100. Qualified in Kings County. Cert. filed with Kings & N. Y. Co. Clerk. Cert. filed with Kings & N. Y. Reg. Offices. Term Expires March 30, 1954.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington 25, D. C.

Appeal No. 33,558

Before the Board of Appeals

Pat. Div. 6

IN RE APPLICATION OF FREDERICK C. BERSWORTH
Serial No. 215,380. Filed March 13, 1951. For Hydroxy Derivatives of Alkylene Polyamines.

AFFIDAVIT-EXHIBIT

Arthur E. Martell, being duly sworn, deposes and says:

That he holds the degree of Doctor of Philosophy in Chemistry from New York University;

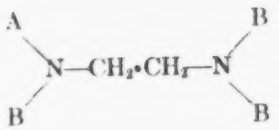
That for the past ten years he has been Professor of Chemistry at Clark University, Worcester, Massachusetts;

Also during the past ten years he has been Chemical Consultant for Frederick C. Bersworth and the Bersworth Chemical Company;

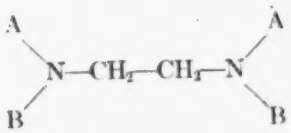
That he is co-author of the book CHEMSITRY OF METAL CHELATE COMPOUNDS, Martell and Calvin, published in September 1952 by Prentice Hall;

That he has studied United States Patent application Serial No. 215,380, the instant case, and related applications filed by F. C. Bersworth, namely Serial No. 491,669 issued as United States Patent 2,407,645; Serial No. 658,893, issued as United States Patent 2,524,218; Serial No. 39,377, abandoned March 20, 1951; Serial No. 39,378, abandoned November 29, 1952.

The applications identified constitute a series and the compounds being claimed in United States application Serial No. 215,380 may be formulated as follows:

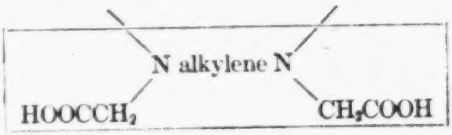



and

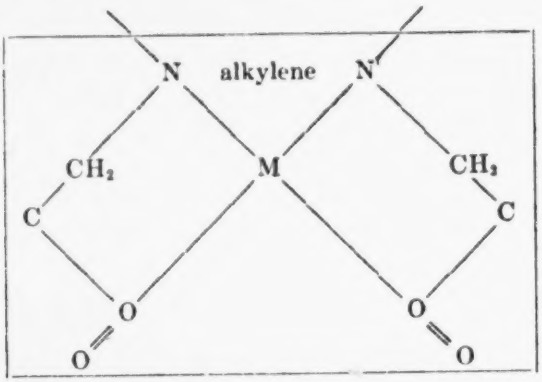



wherein A is a substituent group selected from the group consisting of ethanol and propanol and B is an acetic acid substituent group.

This group of compounds representing diacetic or triacetic acid derivatives of a fundamental ethylene diamine compound have the unusual characteristic that they are exceptionally strong complexing agents by virtue of the fact that they include the following characteristic structural unit:



This structural unit characterizes these compounds, because when the compounds are put in a solution and the solution reacted with a salt of a heavy polyvalent metal, that metal enters into combination with the molecule having this characteristic structural unit to form a compound as follows:




Conditions for the formation of a stable complex of this general form are at their optimum when the N's are so oriented that a group of 5 or 6 membered rings is formed.

This class of compounds being claimed in the instant case has been found to have exceptional utility because the molecule includes alcohol functions attached to the nitrogen atoms, which contribute to the water solubility of the compound. The special alcohol functions provided are due to hydroxyl groups which are substituted on carbon atoms in the $\beta$-position with respect to the nitrogen atom (i. e., 2 carbons between O and N atoms).

The characteristic of the chelate or complex is that it will ionize only very slightly in solution and thus make available traces of the free metal ions as follows:

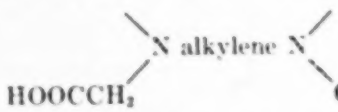
Divalent metal $MA^{2-n} \rightleftarrows M^{+2} + A^{-n}$
Trivalent metal $MA^{3-n} \rightleftarrows M^{+3} \quad A^{-n}$

It has been found that these compounds may be employed to great advantage, for example, in the

control of trace amounts of iron in the soil and in plants growing in such soil. In citrus groves in Florida, a substantial problem in the control of iron chlorosis of citrus trees has arisen, the condition being one which adversely affects the chlorophyll contained in the foliage and the productive capacity of the trees. It has been found that the iron chelates of the compound defined in the instant case are useful in applying appropriate trace amounts of iron to the soil in which these plants are growing to the advantage of the tree in curing the condition and improving its productivity.

Attached hereto and forming a part of this affidavit is a copy of a letter from the Agricultural Experimental Station of the College of Agriculture of the University of Florida reporting some favorable results in the application of iron to citrus trees in the control of iron chlorosis.

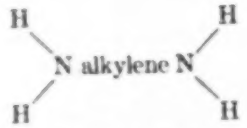
Attached hereto and forming a part of this affidavit is Exhibit B which constitutes an outline of the subject matter which your applicant has had pending in his patent applications before the United States Patent Office, since at least about June 1943. Certain references have been cited in the cases and in order clearly to demonstrate the continuity of the claimed subject matter before the Patent Office, the subject matter of the several related patents and applications has been abstracted and outlined below. The thread of continuity is found in the occurrence of the following characteristic structure in the compounds constituting the essence of the several disclosures:




It should be noted that the characteristic structure is one which involves two carboxyl groups and a pair

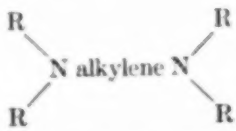
of nitrogen atoms joined through a relatively short carbon chain. Wherever this structural entity occurs in a formula, for clarity in tracing it, it has been blocked in.

It should be noted that in considering ethylene diamine derivatives, the following formulation is evaluated:

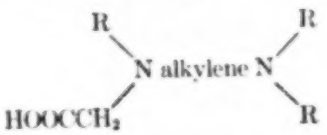



where alkylene is $-CH_2CH_2CH_2-$, $-CH_2CH_2$, or $-CH(CH_3)CH_2$, 1,3– propylene, ethylene, and 1,2– propylene respectively.

A fully substituted ethylene diamine derivative could be considered to be as follows wherein each of the replaceable hydrogens of the nitrogens has been replaced by a radical R:

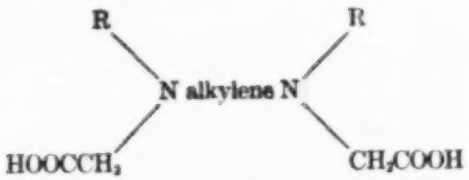



Thus, a monoacetic acid derivative would have the following formulation wherein the R may stand for any alkyl or alkoxy groups.



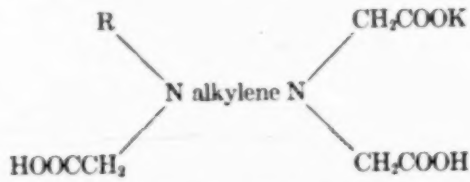

It is clear that in a compound of this form the characteristic structural entity identified as needed for strong chelation does not appear for only a single carboxyl group occurs.

The N N′ diacetic acid derivative would have the following formulation:



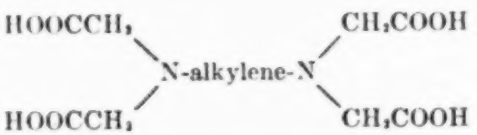

It should be noted that in the N N′ diacetic acid derivative, where a carboxyl group is on each of the nitrogen atoms, the characteristic chelating structure is formed.

The triacetic acid derivative of ethylene diamine would have the following structure:

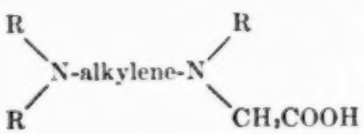



It will be noted here also that the characteristic structural entity including two carboxyl groups attached to the carbon nitrogen chain occurs and powerful chelating action will develop.

The tetraacetic acid derivative would have the following formulation and it has valuable chelating properties:

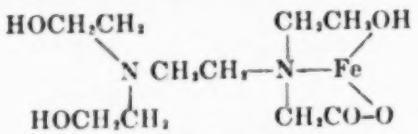



It should be noted that the monoacetic acid derivative which has the following formula




can have chelating properties with conversion of R to $CH_2CH_2OH$.

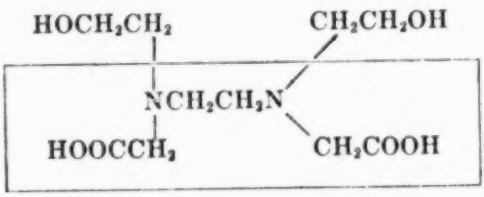
However the chelating power is very weak and is dependent primarily on the carboxyl group and only one adjacent nitrogen atom. For example, with a metal ion such as Fe III the binding occurs to only the carboxyl and the adjacent nitrogen and hydroxyl groups, providing only two chelate rings



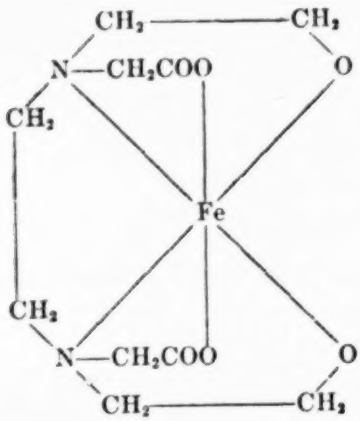

Type A

The resulting chelate rings are not sufficiently stable to prevent the precipitation of ferric hydroxide or phosphate even in moderately acid solutions.

In contrast to this is the very stable chelate structure formed when a carboxyl group is attached to both nitrogens. Thus from

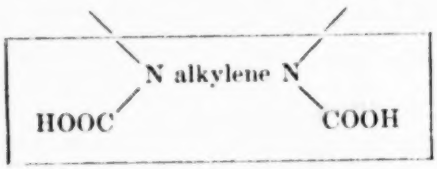



the very stable chelate is formed:



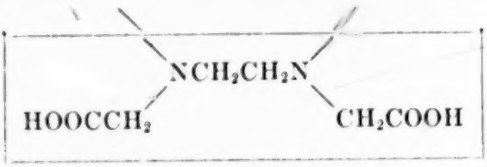

in which the iron is coordinatively saturated, and is not precipitated in acid and alkaline solution as the hydroxide, phosphate, and the like.

Hence it is seen that the coordinating power of the amino nitrogen toward metal such as $Fe^{+3}$ (and also others, such as $Al^{+3}$, $Cr^{+3}$, $Ca^{+2}$, $Mg^{+2}$, $Pb^{+2}$) requires an adjacent acetate group for development. This is obtained only in the compounds having the basic chelate structure outlined above, namely




,

Your applicant has found that the inclusion of an ethanol group on the molecule in addition to the symmetrical group containing the two carboxyl units has a very powerful effect on the chelating properties, particularly on the properties of the chelate compound itself, for it contributes to the solubility of the complex and is especially valuable in the complexing of iron. This property appears only in the mono and di-ethanol derivatives of the ethylene diamine acetic acid derivatives having the characteristic two carboxyl structural entity in the molecule:




In tracing this structure through the series of applications, it is to be noted that in (1) United States Patent 2,407,645 the initial disclosure of the mono-ethanol derivative of ethylene diamine triacetic acid appears.

Copending with this application was (2) United States Patent 2,524,218 and which discloses the diethanol derivatives of ethylene diamine diacetic acid generically.

Copending with that application (2) was United States Patent Application (3) Serial No. 39,377, which contains the claims to the composition of matter divided out of the parent case, United States Patent 2,524,218.

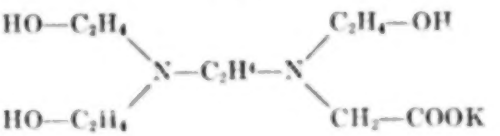
Copending with that case (2) was United States patent application (4) Serial No. 39,378 which contains claims directed to the method of forming the ethanol derivatives.

Copending with United States Patent Application (3) Serial No. 39,377 is United States Patent Application (5) Serial No. 215,380, in which the claims were ultimately restricted to the ethanol and propanol derivatives of ethylene diamine diacetic acid, and triacetic acid.

In order further to illustrate clearly the continuity of this subject matter and the unifying thread, the characteristic structural entity of the pair of carboxyl groups attached to the carbon-nitrogen chain, there is appended hereto a graphical chart wherein certain date lines have been established and time lines have been drawn indicating the period of pendency of specific applications. Connecting each of these pendency lines is a line identifying an illustration of the formula for the compounds being claimed in the said application and an indication wherein the formula defined reads on the subject matter of the instant application.

The principal item of art cited is:

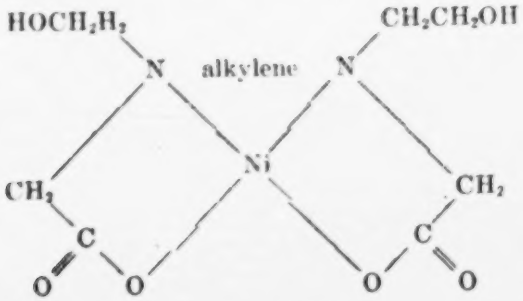
German Patent 642,244 which discloses a variety of compounds of which the Examiner has pointed to the following compound at page 7 thereof:



It will be observed that this compound is an ethylene diamine derivative and may be considered a tri-ethanol derivative of ethylene diamine monoacetic acid. This compound will have some chelating power and will form chelate of the type set forth as type A on page 6 hereof.

It does not display the characteristic structural entity containing the two carboxyl units identified by applicant, which display extreme potency as chelating agents. It further does not disclose the advantage of combining this kind of chelating agent with one or two ethanol groups to attain the controlled solubility of metal complexes formed therewith.

Such transition metals as Ni combine with three groups—2 N's and COOH giving two rings, while with a carboxyl on each N, the metal combines with four to six groups, to give at least the stable structure



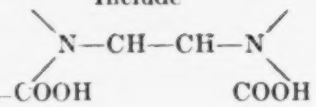

and possibly the hydroxyls (one or both) are also bonded to the metal.

Experimental confirmation of the fact that the compound disclosed in this reference is chemically and characteristically different from that which contains the symmetrical group containing two carboxyl units connected to a nitrogen chain is developed as follows:

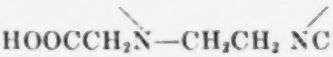
For the reaction $M^{+x} + A^{-n} \rightleftharpoons MA^{+x-n}$ we have the following equilibrium constants, using $Ni^{+2}$ and $Mn^{+2}$ as examples:—

| Chelating Agent | $K_{Ni}^{+2}$ | $K_{Mn}^{+2}$ |
|---|---|---|
| 4 COOH | $10^{18}$ | $10^{13}$ |
| 3 COOH, 1 OH | $10^{16}$ | $10^{12}$ |
| 2 COOH, 2 OH | $10^{15}$ | $10^{11}$ |
| 1 COOH, 3 OH | $10^{10}$ | $10^{6}$ |
| 4 OH | $10^{8}$ | $10^{4}$ |

Include




It is seen that the chelate stability (formation) constants in the table are very high when the basic structure




is present, but the stability falls to a lower level and remains relatively constant for the last two amino acids, which do not have the required structure.

The differences in constants separating the two groups are factors of $10^5$ (100,000 when written out).

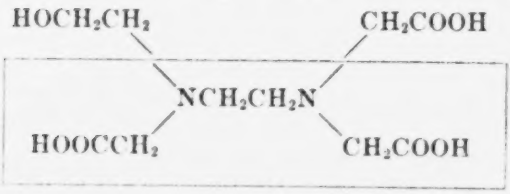
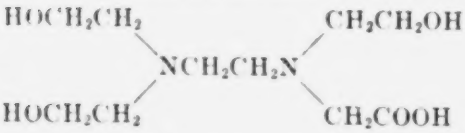
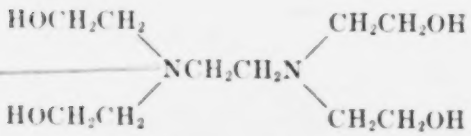
With ferric ion, the effect is even more dramatic. Because of displacement of the hydrogen ion from some of the hydroxyls, the reagents are best compared by the concentration of $Fe^{+3}$ ion in equilbrium at

pH7 with equimolar amounts of excess chelating agent and of the ferric chelate.

For this equilibrium.

$$\text{amino acid anion (at pH7)} + Fe^{+3} \rightleftarrows \text{ferric chelate (at pH7)}$$

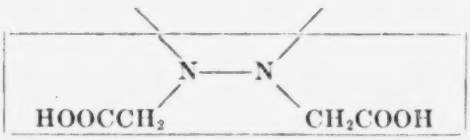
when amino acid anion and ferric chelate are equal we have the following approximate $Fe^{+3}$ levels.

| Amino Acid | Molar Concentration of free $Fe^{+3}$ |
|---|---|
| | $10^{-22}$ |
|  | $<10^{-30}$ |
| | $<10^{-30}$ |
|  Compound identified in German Patent 642,244 | $Fe^{+3}$ Precipitates as Ferric hydroxide (i. e. chelate decomposes by hydrolysis) |
|  | Ferric hydroxide precipitates as above |

A final consideration which points to the subject matter contained in the instant application as inventive may be referred back to the definition or the word "chelate". "Chelate" is a picturesque term

derived from the Greek "chelos", meaning claw. The picture suggested by the term "chelate" is a claw for seizing upon an atom or heavy ion to form a characteristic stable ring compound. An analogy which might be considered is a pair of ice tongs where the claw is symmetrical. Similar to this is a pair of pliers where the claw is symmetrical and a substantial force is developed when properly used.

Applicant has carried through his synthesis of chelating agents, this concept of symmetry based on the characteristic structural entity in two carboxyl groups on the carbon nitrogen chain. The group having the carboxyl groups so arranged on this chain as follows:




It is conceivable and there do occur compounds which have chelating properties which are not symmetrical, but inasmuch as they form complexes of some degree of stability, they are classed generally as chelates. Into this class of compounds can be placed the one disclosed in the German reference. However, chemically, this unsymmetrical material is not only clearly distinguishable, but is self-distinguishing from those compounds which have the characteristic symmetrical chelate forming or claw structure.

ARTHUR E. MARTELL.

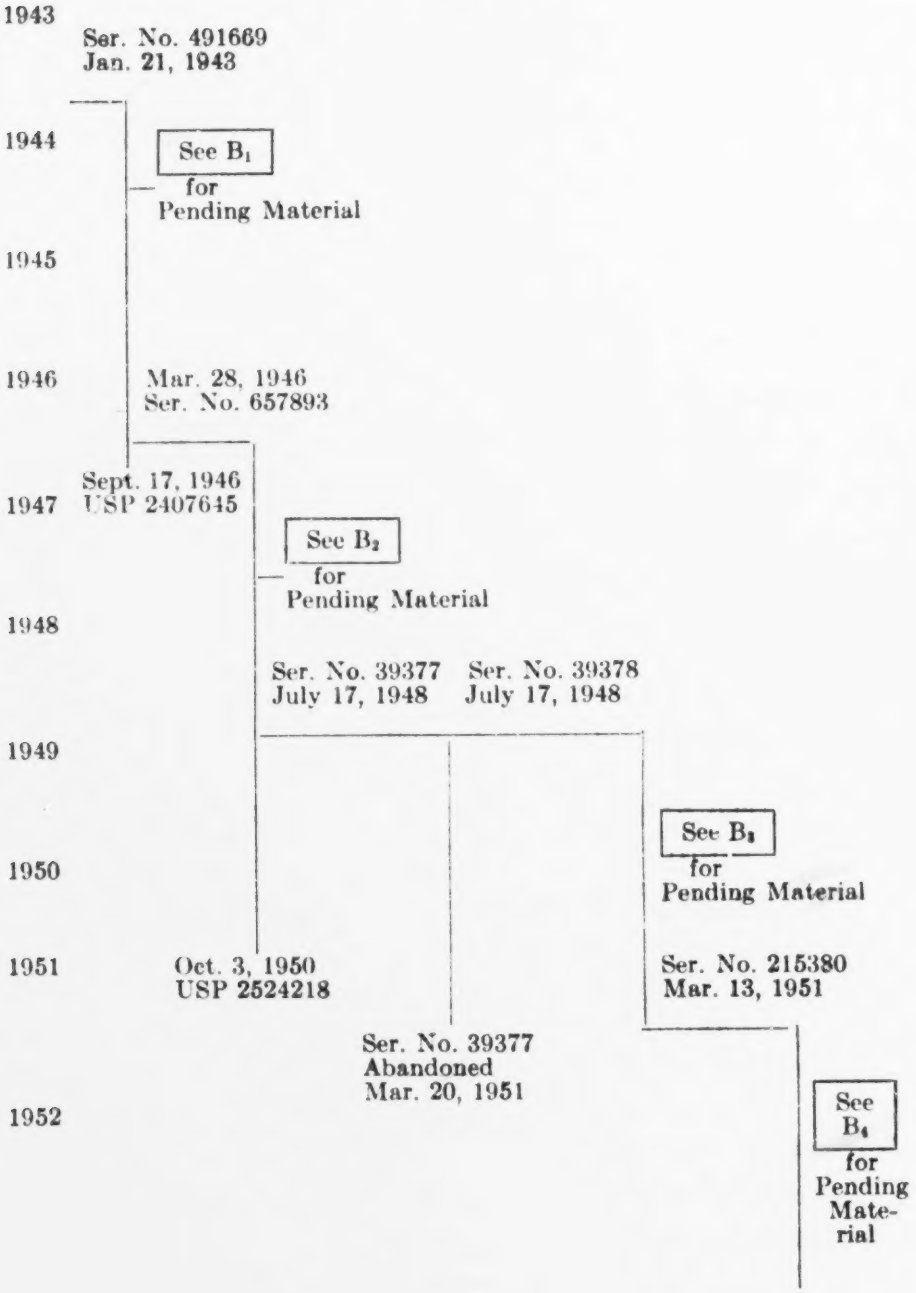
COUNTY OF MIDDLESEX, )
*Town of Framingham, Massachusetts*)

Subscribed and sworn to before me this 12th day of December, 1952.

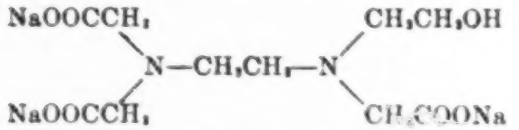
[SEAL] ROSE ERESIAN,
*Notary Public.*

My commission expires May 16, 1958.

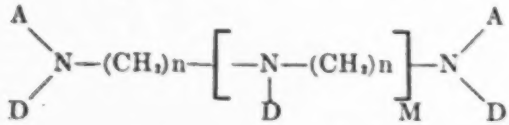
## *Exhibit B*


1943
Ser. No. 491669
Jan. 21, 1943
1944
See B1
for
Pending Material
1945
1946
Mar. 28, 1946
Ser. No. 657893
Sept. 17, 1946
USP 2407645
1947
See B2
for
Pending Material
1948
Ser. No. 39377
July 17, 1948
Ser. No. 39378
July 17, 1948
1949
See B3
for
Pending Material
1950
1951
Oct. 3, 1950
USP 2524218
Ser. No. 215380
Mar. 13, 1951
Ser. No. 39377
Abandoned
Mar. 20, 1951
See
B4
for
Pending
Mate-
rial
1952

U. S. Patent No. 2,407,645, Ser. No. 491,669, filed June 21, 1943 discloses the following compound:



U. S. Patent No. 2,524,218, Ser. No. 657,893, filed Mar. 28, 1946 discloses the following:

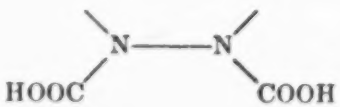


wherein n is either 2 or 3 and m is either zero or a positive integer and wherein D is a member of the group consisting of

$$-CH_2COOH, -CH_2CH_2COOH$$

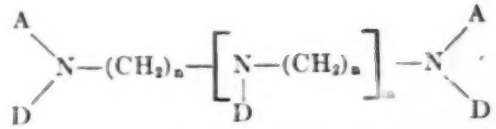
and their alkali metal salts, and where A is a hydrocarbon group containing at least two carbon atoms and having not more than two hydrogen atoms replaced by a member of the group consisting of $-(CH_2)_2COOR$, $-(CH_2)_2COOM$, $-SO_3H$, $-SO_3M$, $-OR$ and $-OR'OR$ wherein R is alkyl or hydrogen, M is an alkali metal, and R′ is alkylene, said groups being spaced at least two carbon atoms from the nitrogen atoms.

Various compounds coming within the scope of that formula are described in the specification. It is to be noted that the

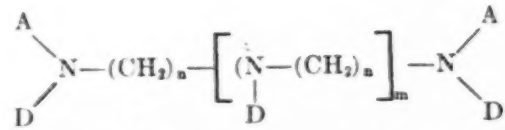


structural entity runs through the disclosure and is found in the above formula when n=2 and M=0 and D=$-CH_2COOH$ Serial No. 39,377, filed July 17, 1948

discloses and claims the following, divided out of Serial No. 657,893:



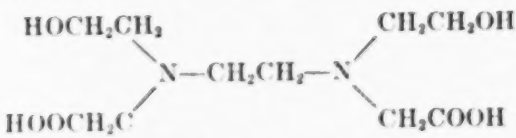
10. The chemical compound conforming to the structural formula:



wherein n is a numeral selected from the group consisting of 2 and 3; m is a value selected from the group consisting of zero and a positive integer; D is a member of the substituent groups consisting of $-CH_2{\cdot}COOH$, $-CH_2{\cdot}CH_2{\cdot}COOH$ and basic metal salts thereof; and A is a hydroxy substituent group selected from the group consisting of mono and poly-hydroxy hydrocarbon compounds.

The characteristic structural entity is in this formula and is clearest in the simplest examples of the series when n=2 and m=0 and D=$-CH_2COOH$.

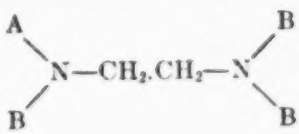
Thus, when n=2, m=0, D=$-CH_2COOH$ and A=$-CH_2CH_2OH$, the formula defines



Serial No. 39378, filed July 17, 1948. This application is directed to methods of preparation.

Serial No. 215,380, filed March 13, 1951 discloses and claims the following:

1. The compound conforming to the formula—



wherein A is a substituent group selected from the group consisting of ethanol and propanol and B is an acetic acid substituent group.

The structural entity is found in this formula as follows:

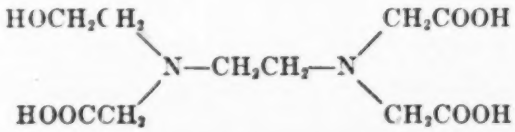



*Exhibit "A"*

Appeal No. 33,558

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 215,380. Filed: March 13, 1951. For: Hydroxy Derivatives of Alkylene Polyamines.

AGRICULTURAL EXPERIMENT STATION

COLLEGE OF AGRICULTURE
UNIVERSITY OF FLORIDA
CITRUS EXPERIMENT STATION

LAKE ALFRED, FLORIDA, *October 27, 1952.*

Dr. FREDERICK C. BERSWORTH,
*Bersworth Chemical Company,*
*Framingham, Massachusetts.*

DEAR FRED: I believe that we told you some time ago that we have some encouraging field results in correcting iron chlorosis on calcerous soils by soil applications of an iron-Versenol Chelate. In fact, we have had positive results from as little as 40 grams of Fe per tree. Since that time, we have put out many more treatments with this material, but evidently results show up more slowly on calcereous soils so we'll have to wait a while longer to see how

they do. We also applied some foliage sprays with both Fe-Versene and Fe-Versenol on trees growing on calcerous soils, and so far the Fe-Versenol looks the better of the two. We have now used all but a small amount of the 30 gallons of 34% Versenol solution which you sent for this work.

We would appreciate your sending another 30 gallons of 34% Versenol solution, or its equivalent in dry form if you have it in dry form, so that we can make additional trials with this material.

We've had over two weeks of cool, wet weather down here, when it rained almost every day and on some days came down like it was being poured from buckets. We got over 3 inches in one flash shower. So far we haven't had a hurricane, although we just had a near miss that went across Cuba.

With best personal regards, I am

Yours very truly,

(S) C. D. LEONARD,
*Assoc. Horticulturist.*

Paper No. 11

Appeal No. 33,558

In the United States Patent Office

BEFORE THE BOARD OF APPEALS

EX PARTE FREDERICK C. BERSWORTH

Application for Patent filed March 13, 1951. Serial No. 215,380. Hydroxy Derivatives of Alkylene Polyamines.

Malcolm N. Rich for appellant.

An affidavit having been filed in this case since the Examiner's answer to appellant's brief was mailed, the application is hereby remanded to the Primary

Examiner for consideration of the merits of the affidavit and the sufficiency of the showing of reasons why it was not earlier presented and for report to the Board of Appeals as to his conclusions thereon.

By Order of the Board of Appeals.

NICHOLAS HAHN,
*Administrative Officer.*

JANUARY 23, 1953.

MALCOLM N. RICH,
*11 Park Place, New York 7, N. Y.*

PAPER NO. 12

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Address only: The Commissioner of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, name of the applicant, and appeal number.

Appeal No. 33558

Before the Board of Appeals

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Ser. No.: 215,380. Filed: March 13, 1951. For: Hydroxy Derivatives of Alkylene Polyamines.

Malcolm N. Rich for Appellant.

EXAMINER'S ANSWER

The applicant has submitted affidavits and the Board of Appeals has remanded the application for report as follows:

(1) As to sufficiency of the showing of reasons why the affidavits were not submitted earlier in the prosecution.

(2) As to the merits of the affidavits in the matter of (1), the sufficiency of reasons as to why the affidavits were not earlier submitted, it appears that the letter from the Agricultural Experiment Station, dated Oct. 27, 1952, concerning work with the compounds claimed, was not available prior to the date of the Examiner's answer.

(3) The affidavits have been carefully considered. The rejection of the second paragraph of paper No. 4 is withdrawn. The other rejections are adhered to and recapitulated as follows:

THE REJECTIONS

Claims 1, 2, 3 and 4 have been rejected as being unpatentable over either of the Bersworth patents No. 8,413,856 and 2,428,353. These patents disclose the general formula where Z is "alkylene" and Y or Y' are "alkylol groups". Patent No. 2,428,353 exemplifies "alkylene" as "ethylene" and exemplifies the "ethanol" substituent, column 3, lines 26 and 27, giving the name of a compound covered by claim 1. Applicant's alleged chain of continuity depends on patent No. 2,524,218, which has not been found to have a disclosure adequate to support the claims at bar. There is a critical difference between the disclosure required to support claims and that necessary for employment as a reference (*In re Stoll et al.*, 603 O. G. 378). To carry the continuity, it is believed that the disclosure must be sufficient to support claims to the subject matter. Applicant's allegation on page 8, third paragraph from the bottom, does not even cover the mono-ethanol derivative *which they say is specifically disclosed in patent No. 2,407,645.* Thus no continuity appears to be alleged as to the compounds of claims 1 and 3. If the disclosure of patent No. 2,524,-

218 is sufficient to support the claims, then by the same reasoning the German patent No. 642,244 anticipates the claims, since this patent has a similar generic disclosure of the subject matter and a specific illustration of the triethanol derivative, applicant covering the other two possible combinations the mono-and diethanol derivatives. It is well-known that this class of materials is useful in the Chelation art, the primary compound in this field being ethylene diamine tetraacetic acid and its salts (Versene) with which applicant, in his enumerated prior patents, considers the compounds at bar as equivalents (In re Borcherdt et al., 665 O. G. 991). The exhibit from the Agricultural Experiment Station indicates Versene (ethylene diamine tetraacetic acid) to be useful in the same application but the Versenol (presumably one of the compounds of the claims) to *look to be a degree* better. This is insufficient for patentability. Other material in the affidavits appear to amount to academic discussion, to opinion or to matters irrelevant to the issue.

The rejections of the first and third paragraphs of text of paper No. 4 are adhered to as noted above.

Respectfully,

H. SURLE,
*Examiner, Division 6.*

MALCOLM N. RICH,
*11 Park Place, New York 7, New York.*

In the United States Patent Office

Appeal No. 33,558

Before the Board of Appeals

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 215,380. Filed: March 13, 1951. For: Hydroxy Derivatives of Alkylene Polyamines.

SUPPLEMENTAL BRIEF

The Examiner having reconsidered the issues raised on appeal in view of the affidavits submitted and having withdrawn the rejection of the second paragraph of Paper No. 4 and recapitulated the other grounds for rejection, this supplemental brief is filed for the purpose of recapitulating appellant's traverse of the recapitulated grounds for final rejection and of answering new grounds for rejection made in the recapitulation.

The Examiner rejects claims 1, 2, 3 and 4 as being *unpatentable* over either prior patent to applicant (Nos. 2,413,856 and 2,428,353).

From the recapitulated remarks of the Examiner this rejection appears to be based primarily upon the finding of fact (page 2, lines 7 to 10 of Statement) that the disclosure of parent patent No. 2,524,218 is inadequate to support these claims.

The Examiner on the authority of *In re Stoll, et al.* (603 O. G. 378; 73 U. S. P. Q. 440; 1947 C. D. 392) states that there is a "critical difference between the disclosure to support claims and that necessary for employment as a reference".

*The Examiner is misconstruing and misapplying this decision as is self-evident from the totally dissimilar facts involved in that case and in the instant*

*case.* There must be some similarity between the facts of the decision and the case in controversy for a decision to be pertinent, persuasive or authoritative. The Board of Appeals therefore is respectfully urged to disregard this decision in the consideration of this appeal. For this decision to apply there must be *prior art* of record. In this case there are prior patents but not prior art of record.

The Board of Appeals is referred to the affidavit of record and particularly to the Chart of Exhibit B depicting the chain of continuity existing between this application and its parent applications. The Board will please note the *issue dates* of the two patents Nos. 2,413,856 and 2,428,353 of *January 7, 1947* and *October 7, 1947* (respectively). The Board will also please note that the parent application of this application has a filing date of January 21, 1943 and that the filing dates of the two prior patents relied on by the Examiner are, respectively, *July 17, 1943* and *June 21, 1943.*

Patents 2,413,856 and 2,428,353 accordingly cannot be considered "prior art" to the instant application as the chain of continuity back to the parent patent 2,407,645 is unbroken. The chart of Exhibit B establishes the fact of application continuity.

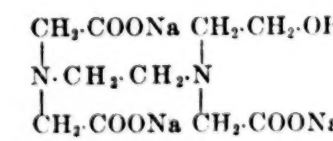
With the above dates in mind, it may be readily recognized from the chart of Exhibit B, that applicant's parent application (Serial No. 491,669), which issued as Patent 2,407,645 on September 17, 1946, was *copending* with both prior patents 2,413,856 and 2,428,353 and was filed *before* these patents were filed and that prior to the issue of either patent 2,413,856 and 2,428,353 applicant had filed application Serial No. 657,893 the parent continuation application to the present application. This application Serial No. 657,893 was pending at the time of issue of its

parent application and either prior patent 2,413,856 and 2,428,353 relied on by the Examiner. Accordingly, under recognized rules of practice *neither prior patent* 2,413,856 and 2,428,353 *is a proper prior art reference* against application Serial No. 657,893 *or against any divisional or continuation application based thereon.*

Under recognized rules of patent law, the only use that can be made of these patents 2,413,856 and 2,428,353 by the Examiner is a rejection based on the rule against *double patenting.*

The claims in issue define a mono and di-ethanol or propanol substituted ethylene diamine tri or di acetic acid compound. The claims of the prior parent patent cover a *process.* The question of double patenting cannot arise between statutorily different patentable inventions.

The Examiner (page 2, lines 15–18) states that the disclosure of applicant's prior patent 2,407,645 lacks a *disclosure* of the mono-ethanol compound. In *direct traverse* of this statement by the Examiner the Board's attention is directed to the disclosure of this patent on page 3 where in the sixth (6th) *species* example of representative products produced by the *process* claimed by this patent is disclosed the N-mono-ethoxy ethylene diamine compound having the formula:



The formula of this disclosed compound in this parent patent conforms to the compound defined by claim 1 on appeal wherein A is ethanol ($CH_2 \cdot CH_2 \cdot OH$).

*In view of this evidence the Examiner's statement on this point is clearly in error.*

Referring now to the continuation application Serial No. 657,893 (now Patent No. 2,524,218) which was *filed prior to the issue date of the parent Patent 2,407,645 and prior to the issue date of either Patent 2,413,856 and 2,428,353,* the Board will note from the affidavit and from the discussion relative to this application of Exhibit B (B–2) that applicant in this parent application disclosed and attempted to claim a *generic* compound which *included* the ethanol and propanol substituted compounds of rejected claims 1 and 2. This is apparent when the A group of the genus compound is construed as defined with respect to the *OR' .OR* group. It is to be noted that the specification of this first continuation application discloses several specific examples of the OR'.OR group compounds.

Out of this generic disclosure of the first continuation (Ser. No. 657,893) two continuation applications (Ser. Nos. 39,377 and 39,378) were filed prior to the issue date of Patent 2,524,218 *and the instant application is a continuation of the application Ser. No. 39,378.*

On the basis of this chain of copending applications it is believed clear that the *species disclosure* of the product invention of the first filed application (now patent 2,407,645) *was not abandoned* on issue of the patent due to its inclusion in the genus disclosure of the first continuation application (Ser. No. 657,893).

The filing of applications Ser. No. 39,377 and Ser. No. 39,378 directed to cover the genus and species of the instant application prior to the issue of patent 2,524,218 completely negatives abandonment of the invention now claimed in the present application which is a continuation of Ser. No. 39,378.

It is respectfully urged that the Examiner is confused as to the law principles involved in the instant

appeal. The rule is well established that the presumption of abandonment of disclosed but unclaimed patentably different inventions in an issued patent is rebutted when the patentee prior to issue of the patent is claiming said unclaimed invention either generically or specifically in a depending application.

The disclosure and original claims in the *first* continuation application Ser. No. 657,893 now issued as Patent 2,524,218 rebut any presumption of abandonment of the ethanol substituted ethylene diamine triacetic acid compound specifically disclosed in the parent patent 2,407,645 and of the genus OH'.OR substituted compound covered by the generic claims of this first filed continuation application.

As the chain of continuity to the instant application is thereafter complete and applicant in the rejected claims is still asserting the species compound first disclosed in the parent patent 2,407,645, *abandonment of this compound on issue of this parent patent is completely rebutted.*

For the reasons stated above the Examiner's rejection based on the false hypothesis of pertinency and applicability of the doctrine of the cited decision to the instant set of facts, is in clear error and should be reversed.

The *sole issue* in this appeal is one of abandonment of a patentably different invention disclosed but not claimed in a prior issued patent to applicant. Applicant is entitled to claim this unclaimed patentably different invention in a continuation application *and once he has done so either generically or specifically the parent patent is thereafter not available to the Examiner as prior art against this invention.* This rule holds as to any timely filed continuation of the first filed continuation.

As the claims of parent patent 2,407,645 are limited to *process* while the claims on appeal are directed to *product* a rejection on the grounds of double patenting cannot be made as between statutorily different classes of patentable inventions.

In view thereof, it appears that the Examiner's rejection based on the German patent also is specious and unavailing. Foreign patents for purposes of anticipation are limited to the *actual* disclosure appearing therein and not by what can be derived by analogy or interpretation therefrom.

The affidavit of record establishes the patentable difference between the triethanol derivative and the mono- and di-ethanol derivatives of the rejected claims and the Examiner has withdrawn this rejection on art.

The Examiner's reference to and rejection based on the *In re Borcherdt, et al.,* decision (page 3 of statement) 665 O. G. 991; 94 U. S. P. Q. 175, is another illustration of faulty reasoning and failure to recognize the precise issue on appeal. This decision also has no applicability to the instant set of facts and "equivalency" is not here in issue.

In this appeal we have the simple issue of the patentability of a disclosed but unclaimed product invention in a parent patent, which is obviously an invention in a *statutorily different class* from the method invention claimed in the parent patent, in a timely filed continuation application derived from the parent patent. No issue of "species-species" relationship exists and no question of "equivalents" exists in the instant issue on appeal to warrant any citation of or reliance on the decision of this *In re Borcherdt, et al.* decision. The disclosures of applicant's prior patents cited and relied on by the Examiner are not available to the Examiner for this ground of rejection. *These patents are not prior art.*

## CONCLUSION

In view of the arguments traversing the rejections by the Examiner it is respectfully submitted that the claims on appeal are allowable to the applicant and that the Examiner should be reversed on all grounds for rejection for the reason that the rule is well established that a patentably different invention which is disclosed but not claimed in a parent patent may be claimed in a continuation application filed prior to the issue date of the parent patent or in a continuation application derived from the first continuation application.

In the instant case the fact that the claims of the parent patent are limited to *process* and the claims of the instant continuation application cover *product* effectively bars any rejection by the Examiner based on the *In re Borcherdt, et al.*, decision.

In the instant case, the fact that applicant's first filed continuation application was filed prior to the issue of either patent 2,413,856 and 2,428,353 effectively removes these prior patents from the prior art and bars a rejection based on the In re Stoll decision.

The fact that none of the claims of the prior patents to applicant of record cover the ethanol and propanol compounds of ethylene diamine di- and tri-acetic acids covered by the claims on appeal bars a rejection based on the rule against double patenting.

In view thereof, a favorable action reversing the Examiner is respectfully solicited.

Respectfully submitted.

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His Attorney.*

MARCH 3, 1953.
MNR: W.

In the United States Patent Office

Division 6

IN RE THE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 215,380. Filed: March 13, 1951. For: Hydroxy Derivatives of Alkylene Polyamines.

POWER OF ATTORNEY AND REVOCATION

Hon. COMMISSIONER OF PATENTS,
*Washington, D. C.*

SIR: The undersigned, owner of the entire right, title and interest in the above entitled application, does hereby appoint Darby & Darby of 405 Lexington Avenue, New York 17, New York, Registration No. 10954, his attorneys, with full power of substitution and revocation, to prosecute this application, to make alterations and amendments therein, to sign the drawings, to receive the patent, and to transact all business in the Patent Office in connection therewith, hereby revoking all previous powers of attorney.

Respectfully,

FREDERICK C. BERSWORTH.

Dated: April 20, 1953.

Accepted May 4, 1953.

ROBERT C. WATSON,
*Commissioner.*

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE

WASHINGTON, *May 5, 1953.*

DARBY & DARBY,
*405 Lexington Ave., New York 17, N. Y.*

Applicant Frederick C. Bersworth. Serial No. 215,380. Filed Mar. 13, 1951. For Hydroxy derivatives of alkylene polyamines. Div. 6.

In this case your power of attorney has been accepted.

Very truly yours,

ROBERT C. WATSON,
*Commissioner of Patents.*

Revoking power of attorney to Malcolm N. Rich, 11 Park Place, New York 7, N. Y.

In the United States Patent Office

Before the Board of Appeals

Div. 6

Appeal No. 33,558

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 215,380. Filed March 13, 1951. For Hydroxy Derivatives of Alkylene Polymines.
400 LEXINGTON AVENUE, NEW YORK 17, N. Y.

*May 12, 1953.*

REQUEST FOR EXTENDED HEARING TIME

Hon. COMMISSIONER OF PATENTS,
*Washington, D. C.*

SIR: Because the subject matter of the application above-identified is of a highly technical nature in a specialized field of organic chemistry, and the history of the case is complicated, it is respectfully requested that the time for oral hearing in the instant case be extended to one hour.

Respectfully submitted.

DARBY & DARBY,
By THOMAS B. GORDON,
*Attorneys for Applicant.*

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE

Paper No. 17

WASHINGTON, *May 21, 1953.*

Address only: The Commisisoner of Patents, Washington, D. C.

DARBY & DARBY,
*405 Lexington, Ave., New York, N. Y.*

NOTICE OF HEARING

The case of Frederick C. Bersworth, Appeal No.: 33,558, Serial No: 215,380, will be heard by the Board of Appeals on the 26th day of January 1954.

The hearings will commence at 9: 30 AM and as soon as the argument in one case is concluded, the succeeding case will be taken up.

The time allowed for argument is thirty minutes.

By special leave, obtained before the argument is commenced, the time may be extended.

Very truly yours,

ROBERT C. WATSON,
*Commissioner of Patents.*

Please refer to appeal number in all communications concerning this case.

Appeal No. 33,558 Paper No. 18

In the United States Patent Office

Before the Board of Appeals

EX PARTE FREDERICK C. BERSWORTH

Application for Patent filed March 13, 1951, Serial No. 215,380. Hydroxy Derivatives of Alkylene Polyamines.

Darby & Darby for appellant.

In response to communication filed May 13, 1953, requesting thirty minutes additional time for the hearing of the above-entitled appeal, please be informed that any additional time required may be granted by the tribunal sitting at the time of the hearing, if it be deemed necessary. The case will appear last on the calendar of hearings (notice of date of hearing herewith). The request is granted to this extent only.

By Order of the Board of Appeals.

NICHOLAS HAHN,
*Administrative Officer.*

MAY 21, 1953.

DARBY & DARBY,
*405 Lexington Ave., New York 17, N. Y.*

Appeal No. 335–58 Paper No. 19

Hearing: January 26, 1954.

In the United States Patent Office

Before the Board of Appeals

EX PARTE FREDERICK C. BERSWORTH

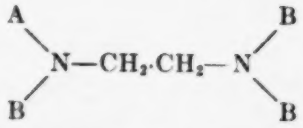
Application for Patent filed March 13, 1951, Serial No. 215,380. Hydroxy Derivatives of Alkylene Polyamines.

Darby & Darby for appellant.

This is an appeal from the action of the Examiner finally rejecting claims 1, 2, 3 and 4, all the claims in the application.

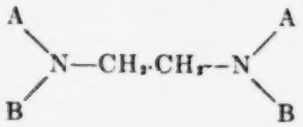
Claims 1 and 2 are representative.

1. The compound conforming to the formula—



wherein A is a substituent group selected from the group consisting of ethanol and propanol and B is an acetic acid substituent group.

2. The compound conforming to the formula—



wherein A is a substituent group selected from the group consisting of ethanol and propanol and B is an acetic acid substituent group.

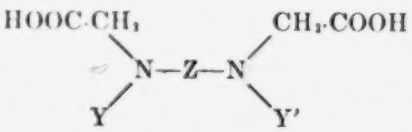
The references relied upon are:

Bersworth—2,413,856, Jan. 7, 1947.

Bersworth—2,428,353, Oct. 7, 1947.

The claims on appeal are to chelating agents which are sufficiently described by the copied claims.

Claims 1, 2, 3 and 4 stand rejected as unpatentable over either of the Bersworth patents Nos. 2,413,856 and 2,428,353. These patents disclose compounds of the general formula



where Z is alkylene and Y and Y′ are alkylol groups. In 2,428,353 the alkylene is disclosed as ethylene, for example, and the alkylol substituent may be ethanol. In column 3, lines 26 and 27 ethylene diamine N,N′-triacetic acid N′-ethanol is disclosed by name. This is one of the compounds covered by the presently appealed claim 1.

There appears to be no dispute on the fact that these patents disclose compounds which anticipate the appealed claims. The argument for patentability is that these patents are not proper references by virtue of the fact that appellant had pending before the Patent Office this subject matter prior to the effective dates of these patents. Appellant traces a chain of continuity back to the parent patent 2,407,645 filed June 21, 1943. Appellant states that the filing of applications Serial Nos. 39,377 and 39,378, which covered the genus and species of the instant application prior to issue of Patent No. 2,524,218, completely negatives abandonment of the invention now claimed in the present application, which is a continuation of Serial No. 39,377.

The alleged chain of continuity depends *inter alia* on Patent No. 2,524,218, which the Examiner has found not to have a disclosure adequate to support the present claims. The Examiner has referred to *In re* Stoll et al, 603 O. G. 378; 1947 C. D. 392; 34 C. C. P. A. 1058; 161 F. (2d) 241; 72 USPQ 440 to support his proposition that there is a critical difference between the disclosure required to support claims and that necessary for employment as a reference. The pertinent disclosure in 2,524,218 relied on for continuity is found in column 1, lines 25 to 46 inclusive. This portion of the patent reproduces a formula with lettered substituents. By selecting certain values and substituents within this disclosure, such as when M is zero, A is a hydrocarbon containing at least two carbon atoms, a hydrogen of which is replaced by OR wherein R is hydrogen, then the compounds now claimed would be reproduced. The disclosure in Patent 2,524,218 does not specifically disclose any of the compounds now claimed hence by the usual standards would not support such claims. Since adequate sup-

port in this patent would be necessary for appellant to overcome the effect of the disclosures in Patents 2,413,856 and 2,428,353 as prior art, it follows that these patents constitute proper references and that the rejection of the Examiner thereon must be sustained. The affidavit of Arthur E. Martell has been reviewed but does not appear to have any bearing upon this ground of rejection.

The decision of the Examiner is affirmed.

E. W. GENIESSE,
*Examiner in Chief,*
N. A. ASP,
*Examiner-in-Chief,*
ARTHUR B. MILLER,
*Examiner-in-Chief (Acting),*
*Board of Appeals.*

FEBRUARY 18, 1954.
DARBY & DARBY,
*405 Lexington Ave., New York 17, N. Y.*

In the United States Patent Office

AFFIDAVIT

STATE OF NEW YORK,
*County of New York, ss:*

Thomas H. Whaley, being duly sworn, deposes and says:

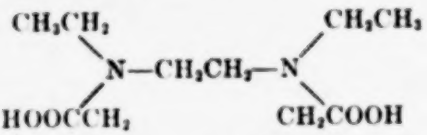
That he holds the degree of Bachelor of Science in Chemical Engineering granted by the University of Colorado in 1937, and that he holds the degree of Master of Science in Chemical Engineering granted by the University of Michigan in 1938;

That for eight years following that period he was in the patent department of the Phillips Petroleum Company and for the past seven years he has been a patent attorney for the Texas Company;

That he is a member of the Bar of the State of Oklahoma, having been admitted in 1945, and that he is registered to practice before the United States Patent Office, Registration 15,028;

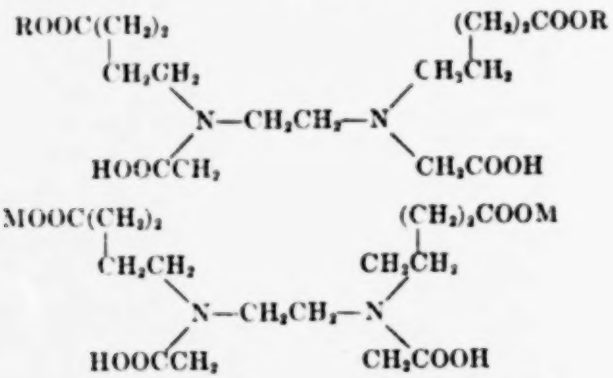
That on Wednesday afternoon, January 27, 1954, he was invited to spend a short period in the office of Mr. Thomas B. Graham, of Darby & Darby, 405 Lexington Avenue, New York, New York, on the representation that Mr. Graham wished to give him a brief chemical test. In the office he was presented with a soft copy of United States Patent 2,524,218, marked at column 1, lines 25 through 45. The single question asked of him was to write out formulas of the compounds which were specifically identified for him by the generic formula there stated using terms as defined. Based only upon the meaning of the generic formula and the definition of the terms therein the following formulas were sketched:

*I*

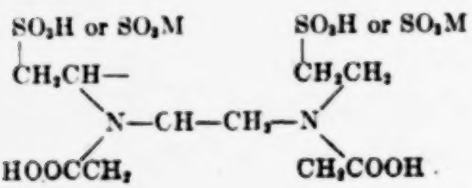


and single substitution in ethyl group gives:

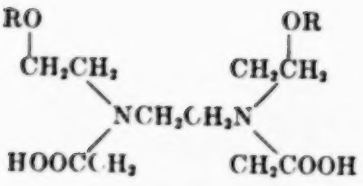
*II*



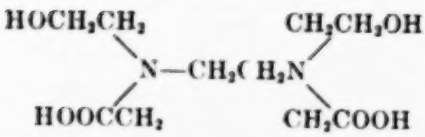
*III*



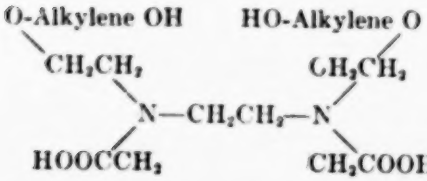
*IV*



and



*V*



That prior to sketching out the formulas above he had no knowledve of the specific subject matter involved or the problem and merely followed fundamental principles of organic chemistry and nomenclature.

That he did not then read the rest of the specification, has not since read the rest of the specification and that after having written out the above formulas, he was told that the reading of the patent was a point at issue in a patent application being prosecuted before the Patent Office.

Thomas H. Whaley,
THOMAS H. WHALEY.

STATE OF NEW YORK,
*County of New York, ss:*

On this 11th day of March, 1954, before me personally came Thomas H. Whaley, to me personally known, and who signed the foregoing instrument in my presence, and duly acknowledged the same to be his free act and deed.

THOMAS A. FLANAGAN,
*Notary Public.*

Notary Public, State of New York. Qualified in Bronx County, No. 03–1246700. Certificates filed in New York and Branx County Clerk's Offices and New York and Bronx Reg's. Offices. Term Expires March 30, 1955.

In the United States Patent Office,

BEFORE THE BOARD OF APPEALS

Appeal No. 335–58

IN RE THE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 215,380. Filed March 13, 1951. For Hydroxy Derivatives of Alkylene Polyamines.

AFFIDAVIT

STATE OF MASSACHUSETTS,
*County of Worcester, ss:*

Arthur E. Martell, being duly sworn, deposes and says:

That he holds the degree of Doctor of Philosophy in Chemistry from New York University;

That for the past eleven years he has been Professor of Chemistry at Clark University, Worcester, Massachusetts;

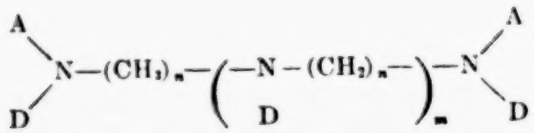
Also during the past eleven years he has been Chemical Consultant for Frederick C. Bersworth and Bersworth Chemical Company;

That he is co-author with Melvin Calvin of the book "Chemistry of Metal Chelate Compounds", published in September 1952 by Prentice Hall;

That at Clark University he carries on a program of active teaching and research, the teaching including classes in elementary and advanced organic chemistry;

That he has examined the specification of U. S. Patent 2,524,218 and reads the disclosure as follows:

The generic formula stated at column 1, lines 25–32 is:



and that the definitions of substituent groups which may be entered into the places designated in the formula may be broken down as follows:

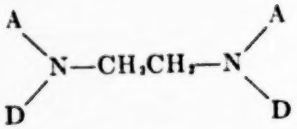
Wherein n is either 2 or 3 and m is either zero or a positive integer and wherein D is a member of the group consisting of $-CH_2COOH$, $-CH_2CH_2COOH$ and their alkali metal salts, and wherein A is a hydrocarbon group containing at least *two carbon atoms,* and having not more than *two hydrogen atoms* replaced by a member of the group consisting of—

$-(CH_2)_2COOR$
$-(CH_2)_2COOM$
$-SO_3H$
$-SO_3M$
$-OR$ and
$-OR'OR$

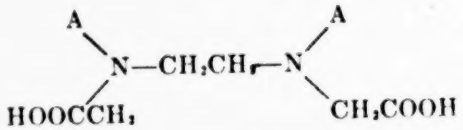
wherein R is alkyl or hydrogen, M is an alkali metal, and R′ is alkylene, said groups being *spaced at least two carbon atoms from the nitrogen atoms.*

In the reading of this general formula it is of basic significance that the A substituent is defined as a hydrocarbon group containing at least two carbon atoms, with no indication of an upper limit on its size. Hence, in determining the specific meaning of the formula the only specific hydrocarbon group identified for the A positions is that containing two carbon atoms or the ethyl group.

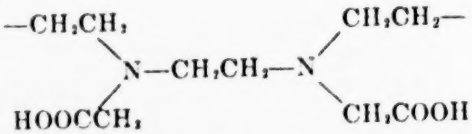
The first step in analyzing the significance of the generic formula, even without resort to the specification, requires that the reader evaluate the structure of the simplest compounds coming within the scope of the definition. In so doing, taking the variables in their order as given and assigning to each the lowest value suggested, we find that when n is 2 and m is 0, the generic formula becomes the following:



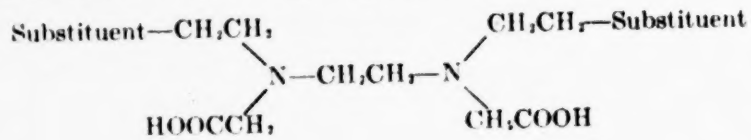
Carrying this to the next step with D as $-CH_2COOH$, the formula becomes the following:



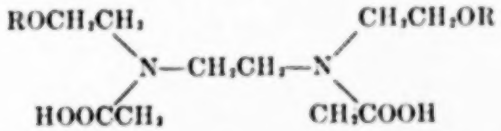
The only remaining variable to be explored is A, and since the only specific identification for the A substituent in the generic definition is the ethyl group, its substitution into the formula produces the following:



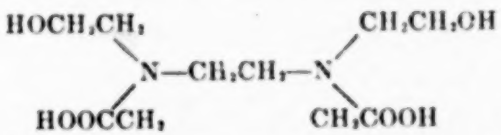
The definition of variables given requires that any substitution on the A group must be spaced at least two carbon atoms from the nitrogen atoms, with the result that the following is produced:



At this point the definition in the generic formula gives the reader the alternative of selecting a hydrophyllic substituent from a group made up of six possibilities. Of these possibilities the OR group is the simplest and the one emphasized in the specification, and evaluating it, the generic formula with the entry of OR in the ethyl groups becomes the following:




and since R may be hydrogen or alkyl, the simplest and only specific form of OR identified is that with the insertion of hydrogen, the diethanol compound



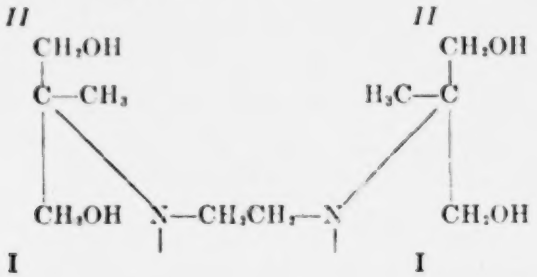

The specification goes on to explore the addition of two OR groups spaced from the nitrogen atom, and also develops the addition of soaplike groups on carbon chains carrying hydroxyl groups.

The form in which the formula is stated is clear and the considerations stated above are developed on the basis of organic chemical nomenclature.

At column 2, lines 28–41 the importance of orienting the hydrophyllic substituent groups in the A positions two carbon atoms or farther out from the nitrogen atoms is pointed out. This is all consistent with the form of the definition of the A group which is stated specifically to be the ethyl group or larger.

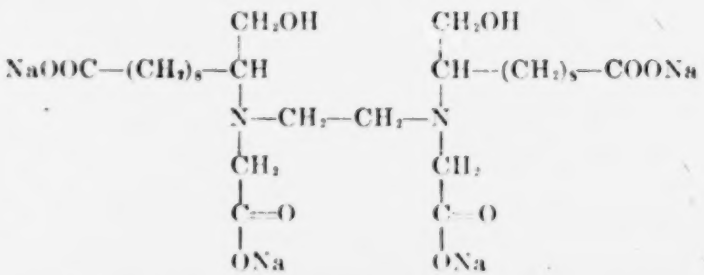
In evaluating which substituents should be used on the chains placed in the A positions, the specification is further illuminating, for at column 4, lines 9–50 emphasis is placed upon the development of the substituted two carbon substituent in the A position. At

line 35–40 the specific compound described is one which is a literal projection of diethanol ethylene diamine diacetic acid, for in the A positions the following groups may be identified:




In the formula drawn above the section identified as I is an ethanol group, the OH being spaced 2 carbon atoms from the nitrogen; that identified as II is also an ethanol group with the OH being spaced 2 carbon atoms from the nitrogen. In other words, that compound is a projection beyond the simplest ethyl derivative which the generic formula dictates in that it shows the attachment of two –OH groups on two carbon atoms, each –OH being removed two carbon atoms from the nitrogen atom.

This projection of the ethyl substitution in the A positions is further illustrated in column 6, lines 6–15 where the following is given




In the formula reproduced above the ethanol group produced by the –OH substitution on the second carbon atom from the nitrogen atom is blocked in. The typographical arrangement selected by applicant also

points to emphasis on the ethanol. Also following the rules for substitution given in the generic formula, the COONa group is substituted on a position farther out from the nitrogen in order to illustrate extremes of positioning of hydrophyllic substituents. The compound reproduced structurally may be looked upon as a projection of substituent groups starting with diethanol ethylenediamine diacetic acid.

Arthur E. Martell.
ARTHUR E. MARTELL.

STATE OF MASSACHUSETTS,
*County of Worcester, ss:*

On this 15th day of March, 1954, before me personally came Arthur E. Martell, to me personally known, and who signed the foregoing instrument in my presence, and duly acknowledged the same to be his free act and deed.

PHILIP H. BREEN,
*Notary Public.*

My commission expires 1/16/59.

In the United States Patent Office

BEFORE THE BOARD OF APPEALS

Appeal No. 335–58

IN RE THE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 215,380. Filed March 13, 1951. For: Hydroxy derivations of alkylene polyamines.

AFFIDAVIT

Thomas B. Graham, being duly sworn, deposes and says:

That he is the Thomas B. Graham identified in the affidavit of Thomas H. Whaley incorporated herein;

That he requested Mr. Whaley to take the test on January 27, 1954, the day after he had participated in the hearing in Appeal 335–58;

That he provided Mr. Whaley with a soft copy of United States Patent 2,524,218, having marked column 1, lines 25–45 with a marginal bracket;

That he simply requested Mr. Whaley to write out specific formulas corresponding to the marked section of the specification;

That he timed Mr. Whaley's sketching of the formulas without advising him that this was being done;

That he observed Mr. Whaley commence sketching formulas after one minute's reading of the text and that the formula identified as IV–B was sketched 4½ minutes after the start of the test.

Thomas B. Graham.
THOMAS B. GRAHAM.

STATE OF NEW YORK,
*County of New York, ss:*

On this 15th day of March, 1954, before me personally came Thomas B. Graham, to me personally known, and who signed the foregoing instrument in my presence, and duly acknowledged the same to be his free act and deed.

Eileen Lamerson.
EILEEN LAMERSON.
*Notary Public.*

Notary Public, State of New York, No. 24–7410100. Qualified in Kings County. Cert. filed with Kings & N. Y. Co. Clerk. Cert. filed with Kings & N. Y. Reg. Offices. Term Expires March 30, 1954.

In the United States Patent Office

BEFORE THE BOARD OF APPEALS

Appeal No. 33,558

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 215,380. Filed March 13, 1951. For: Hydroxy Derivatives of Alkylene Polyamines.

PETITION FOR RECONSIDERATION AND REHEARING AND SUPPORTING BRIEF

Applicant respectfully requests that the Board of Appeals reconsider the decision in Appeal No. 335–58, in view of the following circumstances:

1. The decision is based upon the erroneous premise that U. S. Patent 2,524,218, on which applicant relies for establishing continuity of subject matter before the Patent Office, fails to disclose the composition being claimed in the application on appeal.

2. Certain errors of substance in the Board's Opinion, page 3, the full paragraph, demonstrate the basic disclosure was inaccurately read, which led to a legally incorrect conclusion.

Respectfully submitted.

DARBY & DARBY,
By THOMAS B. GRAHAM,
*Attorneys for Applicant.*

NEW YORK, N. Y., *March 17, 1954.*

BRIEF

PROPOSITIONS OF FACT AND LAW IN SUPPORT OF APPLICANT'S PETITION

1. United States Patent 2,524,218, on which applicant depends for continuity of subject matter discloses

generically and specifically subject matter adequate to support the claims on appeal.

2. Claims of a patent must have adequate support in the specification.

3. Subject matter disclosed, but not claimed in a patent is presumed to be dedicated. The presumption is negatived by timely filing of a continuing application.

*Ex parte Mullen & Mullen,* 50 O. G. 837.
*Ex parte Slayter,* 56 U. S. P. Q. 450.
*Ex parte Spence,* 82 U. S. P. Q. 449.

4. In the preparation of a patent disclosure, an applicant may incorporate published matter by reference to support his own disclosure.

*Graver Tank & Mfg. Co., Inc.* v. *Linde Air Products Co.,* 80 U. S. P. Q. 451, 453.

5. A disclosure which accurately identified a compound is adequate to support a claim to the compound.

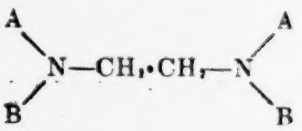
*In re Thacker,* 69 U. S. P. Q. 126.
*Ex parte Dahlen & Zwilgmeyer,* 42 U. S. P. Q. 208.
*Ex parte Moll & Britton,* 81 U. S. P. Q. 447.

ARGUMENT

1. United States Patent 2,524,218, on which applicant depends for continuity of subject matter, discloses generically and specifically subject matter adequate to support the claims on appeal.

This application is before the Board after three requirements of division. The precise subject matter on appeal is the ultimate elected species, claim 2:

"The compound conforming to the formula—



wherein A is a substituent group selected from the group consisting of ethanol and propanol and B is an acetic acid substituent group."

It should be noted carefully that the elected species in this case is the diacetic acid formulated above (Office Action of July 2, 1951 and election of December 28, 1951) and that the substituents for the A group may be ethanol or propanol. As the claim stands it is supported by disclosure of any ethanol or propanol group for the A position.

Continuity of the subject matter of this application is traced from U. S. Patent 2,407,645 (Affidavit of A. E. Martell filed December 31, 1952). The precise history shows that when U. S. Patent 2,407,645 was filed the application contained claims to the method of synthesis and claims to composition of matter. Prompt requirement of division appeared in the Office Action and the election was to proceed with the method claims. The importance of the subject matter is shown by the fact that in applicant's subsequent patents relating to compositions of matter he incorporated this parent patent consistently.

Subsequent to the filing of this parent case, the applications which matured into U. S. Patent 2,428,353 and U. S. Patent 2,413,856 were filed. These cases were all contemporaneous in the Patent Office and issued successively, U. S. Patent 2,407,645 on September 17, 1946, U. S. Patent 2,413,856 on January 7, 1947, and U. S. Patent 2,428,353 on October 7, 1947.

Substantially prior to the issue date of any of these patents, on March 28, 1946, U. S. Patent Application Serial No. 657,893, which matured into U. S. Patent 2,524,218, was filed, containing claims to a class of compounds useful as chelating agents and to a detergent composition. Here again, early in the prosecution of

the case, Office Action required division and the prosecution proceeded on the detergent composition claims.

A reasonable period after the requirement for division U. S. Patent Application Serial No. 735,018, which matured into U. S. Patent 2,532,391, was filed (March 15, 1947) containing claims to compounds divided out of Serial No. 657,893. The divided subject matter directed to the compounds was further subdivided. Specific method and compound claims were presented in application Serial No. 39,377, which expressly included claims to mono- and di-ethanol compounds corresponding to compounds being claimed in this application now on appeal; companion case Serial No. 39,378 contained specific claims to related subject matter. It is the former application, Serial No. 39,377, which was continued into the present application Serial No. 215,380, now on appeal.

The continuity of the subject matter specific to the species being claimed is traceable through the disclosure and claims of the several applications which have been identified.

The presence of the claims in this application is a direct result of successive Patent Office requirements of division and election of species. One species only is now before the Board, namely, the ethanol or propanol substituted dicarboxylic acid of claim 2, quoted above.

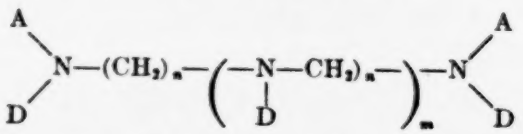
It is in the reading of the disclosure of U. S. Patent 2,524,218 that the Board, like the Examiner, fails to find specific disclosure of the elected species of the instant case.

The formulated disclosure in U. S. Patent 2,524,-218 is cast as a generic expression employing lettered substituents with further detailed discussion in the specification. The reasonable reading of that dis-

closure in the form given logically compels the finding that the patent accurately and specifically discloses the elected species.

It is conventional practice in the Patent Office when new compounds are being claimed to set up a generic definition of the class of compounds in the form of a nucleus which characterizes the compound, and to indicate variants coming within the scope of the invention by employing alphabetical or algebraic symbols at various positions on the nucleus, and to assign to them significance which will lay out the scope of the generic claim. Frequently these generic formulae are referred to as formulae in Markush form and the rules of Markush claims have been applied to them. Such a generic formula necessarily is based upon the disclosure in which it appears and also incorporate inherently the international standard rules of chemical nomenclature. Hence, it is not an arbitrary equation but rather, properly read, a generic formula thus stated is a list by formula and even by name of every compound coming within its scope.

Referring now specifically to the formula given by applicant in his U. S. Patent 2,524,218, it is to be noted that it has the following form:



Evaluating the variables assigned to it a very conspicuous fact immediately appears; namely the value assigned to the A substituent is a hydrocarbon group containing

"at least 2 carbon atoms"

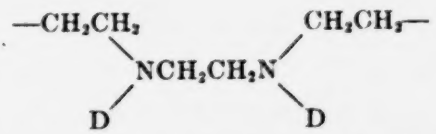
with no assignment of an upper limit. The 2-carbon substituent is the *ethyl* group; hence the only specific

information given about the A group is that it must be at least an ethyl group.

Likewise the value of m is

"zero or a positive integer".

The only specific value of m given is zero. With this bit bit of information the formula immediately becomes:




At this point it is instructive to inspect the Board's reading of the formula, at page 3, lines 15–20:

> * * * By selecting certain values and substituents within this disclosure, such as when M* is zero, A is a hydrocarbon containing at least two carbon atoms, a hydrogen of which is replaced by OR wherein R is hydrogen, then the compounds now claimed would be reproduced.

The Board acknowledges that by "selection" of certain variables the claimed compound is reached. Applicant submits it is not selection but compulsion of the only specific disclosure of the formula which dictates arrival at the compound claimed.

The clauses quoted from the Board's Opinion; namely, m=O, A=hydrocarbon group of 2 carbon atoms, and R=hydrogen, identify the only specific values given to those variables.

The detailed reading of the formula is then traced as follows, for the details given are projections of variations built into the specific ethanol compound disclosed:

---

*The reading of M is obviously in error and should be m.

From U. S. Patent 2,524,218, col. 1, lines 32–45:

| Variable | | Range of Values Specifically Given | |
|---|---|---|---|
| | | *First* | *Ultimate* |
| n | | 2 | 3 |
| m | | 0 | none |
| D | | $-CH_2COOH$ | $-CH_2CH_2COOH$ |
| A | (1) | $-CH_2CH_3$ | none |
| | (2) | $-CH_2CH(CH_2COOH)$ | none |
| | (3) | $-CH_2CH(SO_3H)$ | none |
| | (4) | $-CH_2CH_2OH$ | none |

Four specific forms of A come out of the definitions, the fourth is normal ethanol and its insertion into the formula gives diethanolethylenediaminediacetic acid.

The alternative values for A given in this tabulation are the *specific* ones identified by the generic formula. They are the members of that Markush group.

Thus, reading the formula for the minimum specific disclosure which it gives, one takes the simplest value of A, assigns to it the specific information given about A and immediately thereby identifies a series of compounds, each series beginning with a 2-carbon ethyl group and being characterized by having different substitution in that ethyl group.

Each series thus identified might have been the subject of a separate application, for each member of a Markush group may be considered a genus unto itself. *Ex parte Rutherford,* 63 USPQ 102.

It is to be noted, therefore, that the application being appealed in the instant case has effectively disclaimed many of the variants on the A group described in this generic disclosure. The elected species is that obtained when A is the ethanol or propanol group.

The remainder of the specification of U. S. Patent 2,524,218, logically enough, is essentially an exposition of variations of ethanol substituents in the A position.

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To confirm this reading of the formula the Board's attention is invited to the supplemental affidavit by Dr. Arthur E. Martell, filed herewith and incorporated as part of this argument, wherein the generic formula of U. S. Patent 2,524,218 is objectively stated and it is clearly demonstrated in detail that the inevitable reading of the formula must produce specific disclosure of the diethanolethylene diamine diacetic acid compound.

In further support of such reading of the formula there is incorporated herewith also the affidavit of Mr. Thomas H. Whaley, together with accompanying affidavit of applicant's attorney of record in the case to demonstrate that any person reasonably skilled in the chemical arts need take only that portion of the disclosure appearing in U. S. Patent 2,524,218, column 1, lines 25 to 46 to prepare a list of compounds identifying the several classes called for by applicant.

In recapitulation of the reading of the formula, therefore, it is clear:

1. The "selection" of values in the formula is dictated by the specific disclosure of its context;

2. The Board reads the formula on diethanolethylenediaminediacetic acid;

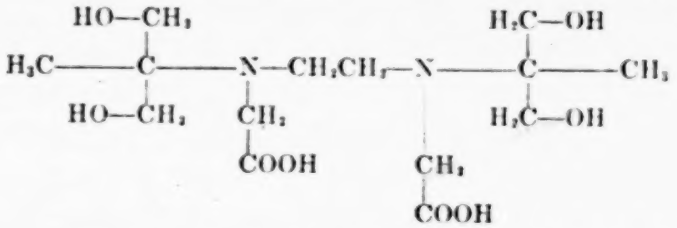
3. Applicant's attorney demonstrates above (page 7) that only four specific forms of A are disclosed, the fourth being ethanol to render the compound diethanolethylenediaminediacetic acid;

4. An eminent scientist and teacher reads the formula to find diethanolethylenediaminediacetic acid in it as the take-off point for elaboration in the accompanying specification;

5. An attorney coming to the disclosure "cold" in five minutes wrote five formulas from the disclosure, four specific ones, one of them being diethanolethylenediaminediacetic acid.

If it can be shown more expressly that the disclosure of U. S. Patent 2,524,218 is adequate to support the claim to the elected species, diethanolethylenediaminediacetic acid, it must be from the patent itself.

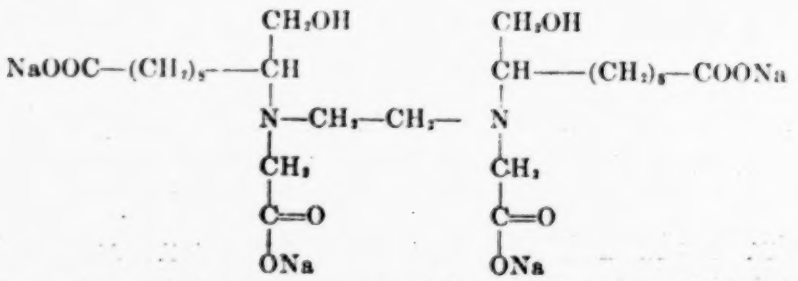
The application as filed contained a generic claim in essentially the language appearing at column 1, lines 25–46; it also contained a specific claim to:



This compound in the blocked in section includes the structure of diethanolethylenediaminediacetic acid (See affidavit of Dr. Martell, filed herewith, page 5. 6).

The compound formulated above could be named systematically as a diethanol derivative. As it stands it is also a propanol" derivative of ethylenediaminediacetic acid (See U. S. Patent 2,524,218, column 4, lines 9–40, and *17*). Hence it comes expressly within the language of claim 2 of the instant application.

U. S. Patent 2,524,218, column 6, lines 5–15 discloses:



The blocked in section includes the structure of diethanolethylenediaminediacetic acid. The compound there formulated could systematically be named as

nonylic acid (9–carbon acid) derivative of diethanolethylenediaminediacetic acid.

Where the generic formula must specifically be read to produce the dicarboxylic acid species employing ethanol or proponol substituents, and the accompanying specification gives two examples solidly within the scope of that elected species, it must be concluded that the disclosure is adequate to support the claim. It is supported by specific formula disclosure of ethanol and propanol compounds corresponding to the claim:

1. The generic formula dictates—diethanolethylenediaminediacetic acid.

2. Column 4, line 17 starts with a propanol (i. e., 1,3-propane-diol) to give—ethylenediamine-N,N′-diacetic acid-N,N′-(1-methyl, 1-methanol) diethanol or ethylenediamine-N,N′-diacetic acid-N,N′-(2-methyl) 1,3-propane-diol.

3. Column 3, lines 6–15, gives the formula for—ethylenediamine-N,N′-diacetic acid—N,N′-(1-nonylic acid) diethanol.

These names are based on the ethylenediaminediacetic acid. Because other definitive names are possible, applicant logically followed the principle: formula ipsa loquitur.

2. Claims of a patent must have adequate support in the specification.

It is an elementary fact of Patent Office procedure that only subject matter which is adequately disclosed in a specification may be covered by the claims of that application. U. S. Patent 2,524,218 contains claim 1 which is directed to a detergent composition consisting of a water-soluble alkali metal fatty acid soap and a certain amount of an alkylene polyamine compound corresponding to the generic formula written, in which the variables are defined in the terms given in the specification. Reading that claim in the

manner which applicant has demonstrated it must be read, namely, by assigning to the A groups the ethyl value, which is specifically disclosed, and then taking the OR or OH substituent, which is also specifically disclosed, the diethanolethylenediaminediacetic acid is derived. It is quite certain that the claim reads upon a detergent composition which would contain from 1 to 5 percent of diethanolethylenediaminediacetic acid within the elected species of the instant application.

The Patent Office apparently considered the specification adequate support for the claim reading specifically on a composition containing diethanolethylenediaminediacetic acid. Is it now consistent to find the specification does not disclose diethanolethylenediaminediacetic acid?

3. Subject matter disclosed but not claimed in a patent is presumed to be dedicated. The presumption is negatived by timely filing of a continuing application.

U. S. Patents 2,413,856 and 2,428,353 have been cited as containing disclosure of the subject matter of the instant application. Specifically, 2,413,856 discloses a generic formula from which a diethanol or monoethanol acetic acid derivative of ethylene diamine can be obtained. The subject matter of the claims, however, relates to plasticized resinous products utilizing esters of these acids, so that the subject matter of the instant application could not have been claimed in this case.

The companion patent 2,428,353 discloses again a generic formula from which mono- or di-ethanol derivatives of the fundamental compound may be obtained, and at column 3, lines 25–30 specifically discloses ethylene diamine N, N′ triacetic acid N′ ethanol, the non-elected species of the instant application.

Based on certain peculiarities of the properties of the esters that class of compounds per se was the subject matter of the patent. The claims applicant is now asserting could not have been asserted in that patent.

Hence, with disclosure of his non-elected species in one of those patents, together with a generic formula which could produce a compound corresponding to the elected species in the instant application, applicant is in the position legally of having disclosed certain subject matter and not having claimed it. The presumption of dedication, of course, arises, but since this application has an effective date long before the issuance of either of these patents, they not only are not prior art, but actually may be part of this disclosure. The Patent Statutes as revised (35 USC § 120) provide that an applicant is to be given the benefit of the filing date of an earlier filed co-pending application if he specifically refers to it in his application, or if his application is amendable to include reference to it. This application is clearly amendable to include reference to any of the prior patents to identify them as parent disclosures.

Applicant's present application has relied upon U. S. Patent 2,524,218 and parent case U. S. Patent 2,407,645. The two patents cited as prior art rely upon U. S. Patent 2,407,645 for disclosure of the method of preparing the compounds and, accordingly, it is perfectly consistent for applicant to incorporate all of this subject matter into his present case by reference. Detailed reference to these two so-called prior art patents has not appeared in this application, but their generic disclosures are useful because of the manner in which the statement of the generic formula has been made. That is, the generic formula as stated clearly indicates that the true concept of applicant's invention as related to the acid forms of his

compounds is to derive chelating agents characterized by having an ethylenediaminediacetic acid structure (affidavit of Dr. A. E. Martell filed December 31, 1952; and election of species, December 28, 1951, Paper No. 3).

Hence, having met and overcome any presumption of abandonment or dedication of subject matter, his case is fully in accordance with the third class of cases discussed in *Ex parte Mullen and Mullen,* 50 O. G. 837, and applicant should be accorded the full significance of his disclosure rather than have fragments of it held against him. Consistent with this view is *Ex parte Slayter,* 56 U. S. P. Q. 450, wherein the Board pointed out with respect to continuity of subject matter, an applicant's only burden is to show his application is "* * * one proceeding as to that concept".

Also see *Ex parte Spence,* 82 U. S. P. Q. 449, and the file of U. S. Patent 2,425,284, Spence, and the consistent line of authorities cited in the record therein.

4. In the preparation of a patent disclosure an applicant may incorporate published matter by reference to support his own disclosure.

It is an elementary fact of patent solicitation that in setting up a disclosure an applicant may begin where the prior art leaves off, describe what is novel, and claim accordingly. Prior art is as much a part of his disclosure as if it were written into it. Where he finds it necessary an applicant may incorporate prior publications into his disclosure by reference. Such practice is not only permissible but expressly encouraged by the Supreme Court. In *Graver Tank & Mfg. Co., Inc.,* v. *Linde Air Products Co.,* 80 USPQ 451, at page 453, the Court had the following to say:

> * * * It would accomplish little to require that claims be separately written if they are

> not to be separately read. While *vain repetition* is no more to be encouraged in patents than in other documents, and claims like other statements may incorporate other matter by reference, their text must be sufficient to "particularly point out and distinctly claim" an *identifiable invention or discovery.* [Emphasis added.]

The quoted passage is of interest for its legal soundness and its substance. Legally it not only permits but encourages precisely what applicant has done.

In cautioning applicants to avoid vain repetition the Court employed a Biblical reference (K. J. V., Gospel, Matthew 6:7) which when read in its context in the opinion is more than a borrowed phrase used for the sake of aptness of the expression. Rather, the use of the language very pointedly epitomizes the lesson of the Linde case—just as the scripture from which the phrase was borrowed admonishes the student to look to the substance of his faith, so the opinion quoted emphatically advises us to look at the substance of a patent disclosure, not merely its literal, technical form.

Applicant has avoided vain repetition in his series of applications, and by incorporation by reference has generic and specific disclosure in his case ample to support his claims.

5. A disclosure which accurately identifies a compound is adequate to support a claim to the compound.

A frequently quoted decision which advocates as well as Examiners must live with is *In re Stoll,* 73 USPQ 440, which the Board, with the Examiner, has relied upon for the proposition that a critical difference exists between disclosure required to support claims and that necessary for its use as a reference. The specific factual matter in that decision concerns

the preparation of an organic compound of biological origin and the rule of the case was that, because the prior art showed hydrogenation of a composition corresponding to that of applicant's starting material, it must have produced the hydrogenated product which he was attempting to claim. In that case the applicant was attempting to dismiss printed subject matter of record as part of the prior art in order to justify the allowance of claims. Precisely how the decision applies in applicant's case has not been shown by the Examiner. Applicant has demonstrated in detail that his disclosure inherently and inevitably requires that it be read to include the formula of the compound he claims. In fact the generic formula given by applicant, by identifying only the first members of a small number of series of compounds without stating upper limits for those series, could be said to disclose little else. Inherently, expressly and logically the formula disclosed in U. S. Patent 2,524,218 lists that which applicant asserts in his claims. It can be conceded that a detailed disclosure is needed to support a claim, but that is precisely what applicant has, for the very specific subject matter of the specification of U. S. Patent 2,524,218 is an exposition of such detail.

In support of applicant's view that the disclosure in U. S. Patent 2,524,218 is adequate to support his claims, the attention of the Board is invited to *In re Thacker,* 69 USPQ 126, where the problem facing that applicant was analogous to the problem facing the applicant here. In that case, to go behind a given reference, the applicant had to find in one of his earlier applications disclosure of the material he was attempting to claim. The earlier application did not name the compound being formed but had expressly set forth each of the steps of the process for making

it. The applicant was conceded his date, and his claims allowed.

Transferring that constructive approach to the instant problem, inevitable logical reading of applicant's disclosure in U. S. Patent 2,524,218, as is amply demonstrated by the affidavits accompanying this brief, clearly demonstrates that inherent in the disclosure is the matter applicant now claims. It not only appears in the patent, but it was considered sufficient to support a claim in that patent reading on a composition containing that compound.

If more authority be needed for support of applicant's reading of disclosure of the form given in his U. S. Patent 2,524,218, the Board's attention is invited to *Ex parte Dahlen and Zwilgmeyer,* 42 USPQ 208. There the Board approved the allowance of specific claims coming within a genus stated in the algebraic form used by applicant expressly on the ground that such designation of species is adequate.

In Ex parte *Moll and Britton,* 81 U. S. P. Q. 447, the Board of Appeals in refusing the applicant's claims of Markush form in diminishing scope emphasized that a Markush claim must be read to include "* * * the group of complete compounds that results from the substitution of the various members of the symbol groups in the compound defined by the structural formula * * *".

This is all applicant has asked the Board to do. When applicant's disclosure in U. S. Patent 2,524,218 is so read in the light of the specific definitions of groups there given, it is inevitable that the patent must be found to have disclosure adequate to support the claims at bar.

## Conclusion

An applicant for a patent is charged with knowledge of the prior art. A corollary of this is to concede to him knowledge he has disclosed in his prior patents and co-pending related applications and give him the benefit of that knowledge. In this application, in full accordance with principles set out in the cases cited, applicant has disclosed:

1. An identifiable invention,
2. Particularly pointed out,
3. Employing information incorporated into his case by reference,
4. In a single proceeding as to that invention.

The Board is accordingly respectfully requested to reconsider its decision of February 18, 1954, and grant applicant a rehearing or a favorable action on the claim on appeal.

Respectfully submitted,

Darby & Darby,
By Thomas J. Graham,
*Attorneys for Applicant.*

Appeal No. 335–58 Paper No. 22

In the United States Patent Office

Before the Board of Appeals

Ex parte Frederick C. Bersworth

Application for Patent filed March 13, 1951, Serial No. 215,380. Hydroxy Derivatives of Alkylene Polyamines.

Darby and Darby for appellant.

## On Petition for Reconsideration

A petition is presented requesting a rehearing and a reconsideration of our decision of February 18, 1954 wherein we affirmed the Examiner in refusing to allow claims 1, 2, 3 and 4. Our crowded docket does not ordinarily permit rehearings and we do not grant them unless there are special circumstances to warrant it. We see no basis for a rehearing in this case, hence the petition will be treated as one for reconsideration.

The petition under point 1 takes issue with our finding that U. S. Patent 2,524,218 does not specifically disclose any of the compounds now claimed and consequently does not provide proper support for such claims. In a lengthy petition accompanied by affidavits appellant seeks to show that with the broad formula of this patent before him and the possible substituents named one would be *compelled* to arrive at the compound now claimed. This premise seems based on the assumption that one would start substituting the simplest possible radicals in the formula and in doing this would inevitably come up with the compound of the present claims. Separate affidavits by Arthur E. Martell and by Thomas H. Whaley have been submitted in an effort to substantiate this contention. The Martell affidavit is summarized by appellant as establishing that a scientist reads the formula "to find diethanolethylene-diaminediacetic acid in it as the take-off point for elaboration". The Whaley affidavit was submitted as evidence that an "attorney coming to the disclosure 'cold' in five minutes wrote five formulas" therefrom

four of which were specific and one being diethanol-ethylenediaminediacetic acid.

The experiments represented by the affidavits may be of interest as mental tests applied to the problem of what likely compounds are embraced by the broad formula in question. Possibly the same tests could be applied to German Patent 642,244. As stated by the Examiner in his Answer of February 13, 1953, Paper No. 12, page 2

> If the disclosure of patent No. 2,524,218 is sufficient to support the claims, then by the same reasoning the German patent No. 642,244 anticipates the claims, since this patent has a similar generic disclosure of the subject matter and a specific illustration of the triethanol derivative, applicant covering the other two possible combinations the mono-and diethanol derivatives.

It should be obvious that these tests and affidavits cannot serve to supply any specific disclosure of the compounds now claimed to Patent No. 2,524,218. Lacking such disclosure we are unable to see how the patent can form proper support for the present claims. The Stoll et al decision appears controlling here. With the alleged chain of continuity broken, Patents 2,413,856 and 2,428,353 become statutory bars. It is therefore unnecessary to consider the question of dedication, point 3 page 13 of the petition. Points 2, 4 and 5 of the petition are merely statements of the obvious such as "Claims of a patent must have adequate support in the specification."

The petition has been considered but is denied in respect to making any change in our decision.

E. W. GENIESSE,
*Examiner-in-Chief,*
N. A. ASP,
*Examiner-in-Chief,*
ARTHUR B. MILLER,
*Examiner-in-Chief*
*(Acting),*
*Board of Appeals.*

MARCH 31, 1954.
DARBY AND DARBY,
*405 Lexington Ave., New York 17, N. Y.*

In the United States Patent Office

BEFORE THE BOARD OF APPEALS

Appeal No. 335–58

IN RE THE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 215,380. Filed March 13, 1951. For: Hydroxy Derivatives of Alkylene Polyamines.

RENEWED PETITION FOR RECONSIDERATION AND REHEARING

Applicant respectfully requests that the Board of Appeals reconsider the decision in Appeal No. 335–58 and its decision dated March 31, 1954, on applicant's petition for reconsideration, because they are based on erroneous premises, summarized and answered as follows:

IN THE OPINION OF THE BOARD
MARCH 31, 1954

I

Page 2, lines 5-8: "* * * This premise seems based on the assumption that one would start substituting the simplest possible radicals in the formula and in doing this would inevitably come up with the compound of the present claims."

II

Page 2, lines 11-14: "* * * The Martell affidavit is summarized by appellant as establishing that a scientist reads the formula 'to find diethanolethylene - diaminediacetic acid in it as the take-off point for elaboration' ".

III

Page 2, lines 14-18: "* * * The Whaley affidavit was submitted as evidence that an "attorney coming to the disclosure 'cold' in five minutes wrote five formulas" therefrom four of which were specific and one being diethanolethylenediaminediactic acid."

APPLICANT'S ANSWER

I

Applicant's premise that the generic formula of United States Patent No. 2,524,218 compels that it be read to disclose diethanolethylenediaminediacetic acid is not based on any assumption. Rather it is based upon the positive assertion of the patent contained in specific disclosure, at col. 1, lines 38–40 and 43–45. At that point in the specification that patent discloses the ethanoldiacetic acid compound and expressly tells the reader to start there and carry the disclosure in the indicated direction. Further express disclosure of ethanol derivatives of ethylenediaminediacetic acid appears at col. 4, lines 35–40 by formula; and at col. 6, lines 6–14 by formula.

II

The Martell affidavit is a demonstration of how one skilled in the art reads the disclosure. It should be noted from pages 5 and 6 of that affidavit that the disclosure is read as being a detailed exposition of the chemistry of diethanolethylenediaminediacetic acid. It is expressly pointed out in the affidavit, page 5, line 14 et seq. and page 6, line 6 et seq., that the compounds expressly described in the specification are diethanolethylendiaminediacetic acids.

III

The Whaley affidavit further demonstrates that the formula in United States patent 2,524,218, dictates ethanol derivatives of diethanolethylenediaminediacetic acid. The form in which the compounds are written in the affidavit emphatically points out that the compounds defined are derivatives of the two car-

| IN THE OPINION OF THE BOARD MARCH 31, 1954 | APPLICANT'S ANSWER |
| --- | --- |
| III | III |
| | bon substituent and the specific one readable directly from the generic formula is diethanolethylenediaminediacetic acid. |
| IV | IV |
| Page 2, line 33, Page 3, line 1: "• • • It should be obvious that these tests and affidavits cannot serve to supply any specific disclosure of the compounds now claimed to Patent No. 2,524,218." | It should be obvious to the Board that this reading of the specification and these affidavits are not an attempt to supply disclosure, but rather are merely an emphatic demonstration that the disclosure stands in the patent. |
| V | V |
| Page 3, lines 1-3: • • • "Lacking such disclosure we are unable to see how the patent can form proper support for the present claims." | Disclosure of the compounds defined in the elected claim is not lacking. It stands expressed by formula at three points in the specification and in claim 1 of U. S. Patent 2,524,218. |

The reading of a generic formula whether it occur in an English text, or any other, can be worked out in accordance with the international rules of nomenclature for systematic identification of chemical compounds; just as formulas are picture representations of the chemical constitution of compounds, the rules of nomenclature constitute an international code for reducing these formulas to word pictures. These formulas, whether they be written in the pictorial form or in the code form, are addressed to those skilled in the art and their sufficiency as disclosure is to be tested by what they tell those skilled in the art. For a recent expression of opinion concerning sufficiency of disclosure, the attention of the Board is invited to *Stauffer Chemical Co.* v. *Watson, Comr. Pats.*, 100 U. S. P. Q. 209, 210 (1954):

> The specification of a patent is not addressed to the public generally, but rather to those persons skilled in the art, and therefore, such things which are familiar to them need not be

pointed out in detail. Any description which is sufficient to apprise those skilled in the art of the definite features of the invention and which gives notice to others of what the patent claims as a monopoly is sufficiently definite to sustain the patent. The disclosure in the instant case is sufficient to enable one skilled in the art to practice the invention.

At page 2, lines 29 to 32 of the Board's opinion of March 31, 1954, the Board adopts the elastic type rejection commonly met in the Examining Divisions, namely, that if rejection A is insufficient, then rejection B must be sufficient. The German patent was cited early in the prosecution. It was technically distinguished. This was accepted by the Examiner. The Martell affidavit of December 31, 1952 expounds the distinctions in detail. The brief filed by applicant's prior attorney, March 3, 1953, p. 6, made reference to the rule of law which states that a foreign patent is good for only what it expressly shows. At the hearing, the Board accepted the statement that the generic formulas disclosed in that German patent do not read directly on the compounds being claimed.

The resurgence of German patent No. 642,244, at this point, amounts to raising a new ground of rejection, or resurrecting a former ground of rejection in a form such that it is effectively a new ground of rejection, and applicant's case should therefore be remanded to the Examiner for full development of this issue.

Accordingly, to clarify the record of this case for further appeal, applicant specifically requests in the alternative:

1. That the Board's affirmance of the Examiner be reversed and applicant's claims be allowed; or

2. A rehearing be granted so that the reading of these disclosures in applicant's U. S. patent 2,524,218

and German patent No. 642,244, may be thoroughly demonstrated to the Board; or

3. That the application be remanded to the Examiner for clarification of the record.

Respectfully submitted.

DARBY & DARBY,
THOMAS E. GRAHAM,
*Attorneys for Applicant.*

_____ _____,
*405 Lexington Avenue,*
*New York 17, N. Y.*
APRIL 2, 1954.

In the United States Patent Office

BEFORE THE BOARD OF APPEALS

Appeal No. 335–58

IN RE THE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 215,380. Filed: March 13, 1951. For: Hydroxy Derivatives of Alkylene Polyamines.

RENEWED PETITION FOR RECONSIDERATION AND REHEARING

In the document captioned as above, dated April 2, 1954, at New York, kindly amend page 2, column headed "Applicant's Answer", paragraph III, line 5, by cancelling "diethanol-".

Respectfully submitted.

DARBY & DARBY,
By HOWARD E. GRAHAM,
*Attorneys for Applicant.*

_____ _____,
*405 Lexington Avenue, New York 17, N. Y.*
APRIL 3, 1954.

Appeal No. 335–58 Paper No. 24

In the United States Patent Office

Before the Board of Appeals

Ex Parte Frederick C. Bersworth

Application for Patent filed March 13, 1951, Serial No. 215,380. Hydroxy Derivatives of Alkylene Polyamines.

Darby and Darby for appellant.

Appellant has filed, on April 5, 1954, a further request for reconsideration of the Board's decision of February 18, 1954.

Since this request comes more than thirty (30) days from the date of the decision, the Board must refuse to act thereon, according to Rule 197 (January 1, 1953) amended March 3, 1954, which states that

> Any request or petition for rehearing or reconsideration, or modification of the decision, must be filed within thirty days from the date of the original decision, unless that decision is so modified as to become, in effect, a new decision, and the Board of Appeals so states.

The file is therefore returned to the Primary Examiner.

By Order of the Board of Appeals.

Nicholas Hahn,
*Administrative Officer.*

April 12, 1954.

Darby and Darby,
*405 Lexington Ave., New York 17, N. Y.*

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Address only The Commissioner of Patents, Washington 25, D. C.

RE: FREDERICK C. BERSWORTH

Serial No. 215,380. Filed March 13, 1951. For: Hydroxy Derivatives of Alkylene Polyamines.

NOTICE OF CIVIL ACTION UNDER 35 U. S. C. 145

A civil action under 35 U. S. C. 145, entitled *Frederick C. Bersworth* v. *Robert C. Watson, Commissioner of Patents*, Civil Action No. 1637–54, involving this application was filed on April 19, 1954, in the United States District Court for the District of Columbia.

E. L. REYNOLDS,
*Solicitor.*

215380

PAPER NO. 4/13/54

MAR 21 1951

DIVISION 6

No. 53

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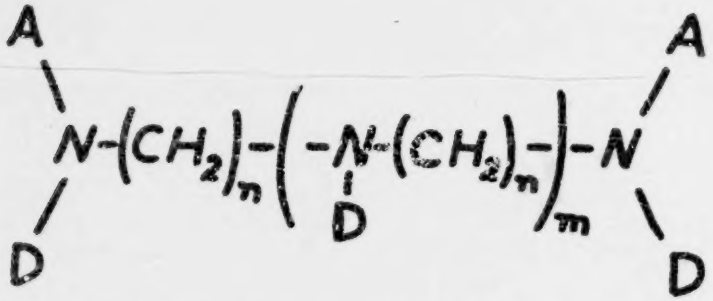
*Plaintiffs' Exhibit No. 13, C. A. 1637–54*

**CASE – C**

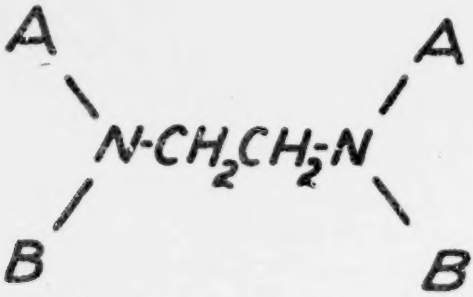
S.N. 215380




*Plaintiffs' Exhibit No. 14, C. A. 1637–54*

**CASE-C**

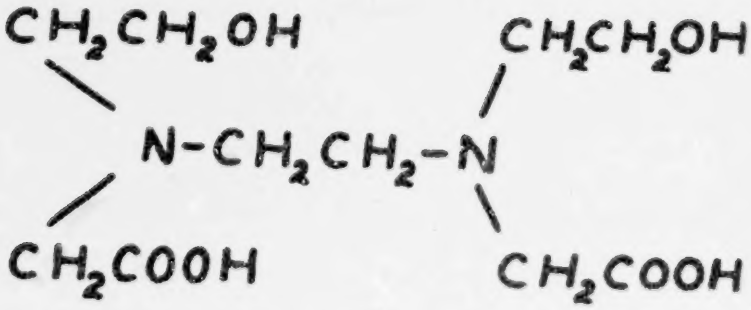
S.N 215380




*Plaintiffs' Exhibit No. 15, C. A. 1637–54*

**CASE – C**

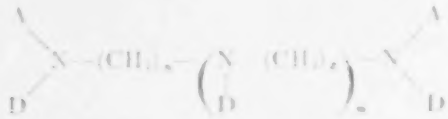
S.N. 215,380



*Plaintiffs' Exhibit No. 16*

I Claim:

1. A compound having the formula



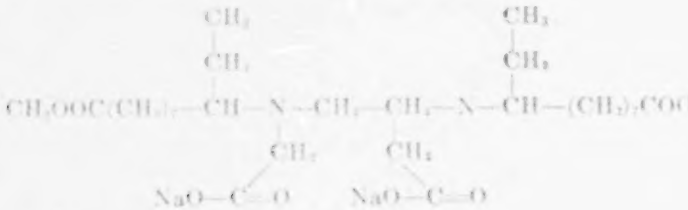
wherein n is a positive whole number from 2 to 3 and m is zero or a positive integer and wherein D is a member of the group consisting of $-CH_2COOH$, $-CH_2CH_2COOH$, and their alkali metal salts, and wherein A is a hydrocarbon group containing at least two carbon atoms and having not more than two hydrogen atoms replaced by a radical of the group consisting of $-(CH_2)_2COOR$, $-(CH_2)_2COOM$, $SO_3H$, $-SO_3M$, $-OR$, and $-OR'OR$ wherein R is a member of the group consisting of alkyl and hydrogen, M is an alkali metal, and R' is alkylene, each said radical being spaced at least two carbon atoms from the nitrogen atoms.

2. A compound according to Claim 1 wherein D is the radical $CH_2COOR$, R being hydrogen or alkali metal.

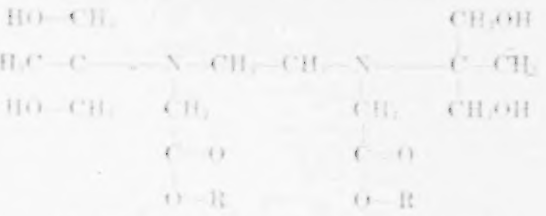
3. A compound according to Claim 1 wherein the hydrocarbon groups are alkyl groups.

4. A compound according to Claim 1 wherein the hydrocarbon groups are cyclic groups.

5. A compound having the formula

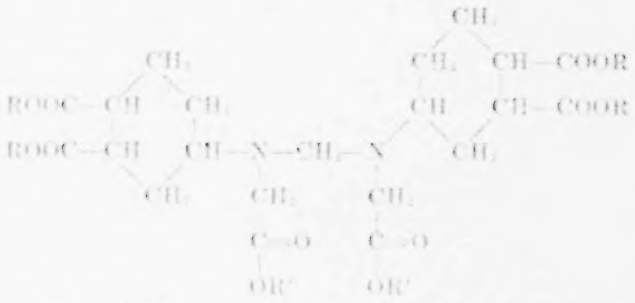


6. A compound having the formula



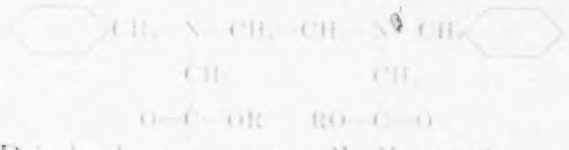
wherein R is hydrogen or an alkali metal.

7. A compound having the formula



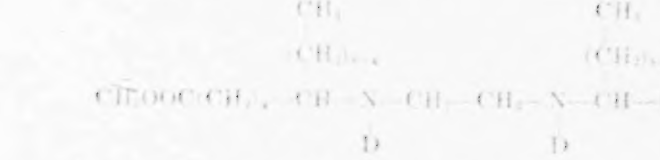
wherein R is hydrogen or an alkyl group and R′ is hydrogen or an alkali metal.

8. A compound having the formula



wherein R is hydrogen or an alkali metal.

9. A compound having the formula



wherein 7 is a positive integer between 2 and 8, inclusive, and D is a member of the group consisting of $-CH_2COOH$, $-CH_2CH_2COOH$, and their alkali metal salts.

Signed at Verona, New Jersey, this 7th day of March 1947.

FREDERICK C. BERSWORTH.

## Oath

State of New Jersey, *County of Essex*, ss:

Frederick C. Bersworth, the above named petitioner, being duly sworn, deposes and says that he is a citizen of the United States, and a resident of Verona in the County of Essex and State of New Jersey; that he verily believes himself to be the original, first, and sole inventor of the invention or discovery in Alkylene Polyamine Derivatives described and claimed in the annexed specification; that he does not know and does not believe that the same was ever known or used before his invention or discovery thereof, or patented or described in any printed publication in any country before his invention or discovery thereof, or in public use or on sale in the United States for more than one year prior to application Serial No. 657,893, filed March 28, 1946 (of which this is in part a continuation) as to subject matter therein, nor more than one year prior to this application, as to the additional subject matter herein; that said invention has not been patented in any country foreign to the United States on an application filed by him or his legal representatives or assigns more than twelve months prior to this application; and that no application for patent on said invention has been filed by him or his representatives or assigns in any country foreign to the United States.

Frederick C. Bersworth.

Subscribed and sworn to before me this 7th day of March 1947.

Herkit M. Appleton,
*Notary Public.*

Notary Public of New Jersey. My Commission Expires Apr. 15, 1951.

In the United States Patent Office

Div. 6. Elmwood, Massachusetts

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Ser. No. 735,018. Filed March 15, 1947 For: Alkylene Polyamine Derivatives.

HON. COMMISSIONER OF PATENTS,
*Washington, D. C.*

SIR: Please amend the above-entitled application as follows:

Claim 1, first line after the formula, cancel "either 2 or 3 and m is either" and insert—a positive whole number from 2 to 3 and m is—

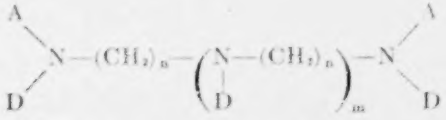
fifth line after the formula, cancel "member" and insert—radical—;

eighth line after the formula, cancel "said groups" and insert—each said radical—.

Cancel claims 5, 7, 8, and 9.

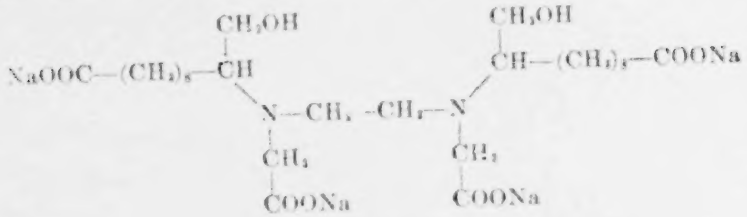
Add the following claims:

10. A compound having the formula

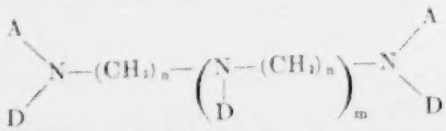


wherein n is a positive whole number from 2 to 3 and m is zero or a positive integer and wherein D is a member of the group consisting of $-CH_2COOH$, $-CH_2CH_2COOH$, and their alkali metal salts, and wherein A is a hydrocarbon group containing at least two carbon atoms and having from one to two hydrogen atoms replaced by the radical $>OH$, each said radical being spaced at least two carbon atoms from the nitrogen atoms.

11. A compound having the formula

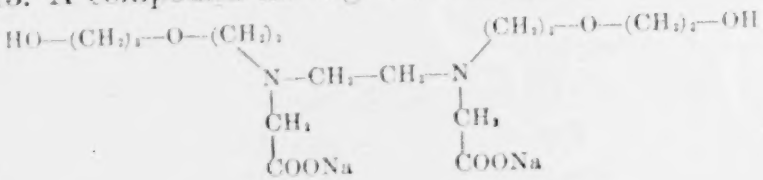


12. A compound having the formula



wherein n is a positive whole number from 2 to 3 and m is zero or a positive integer and wherein D is a member of the group consisting of $-CH_2COOH$, $-CH_2CH_2COOH$, and their alkali metal salts, and wherein A is a hydrocarbon group containing at least two carbon atoms and having from one to two hydrogen atoms replaced by the radical $-OR'OH$ and R′ is alkylene, each said radical being spaced at least two carbon atoms from the nitrogen atoms.

13. A compound having the formula



## Remarks

Claims 5, 7, 8, and 9 have been cancelled because they are substantial duplicates of claims presented in application Ser. No. 737,725 filed March 27, 1947 by applicant.

Claim 1 has been amended slightly to make it clearer.

New claims 10, 11, 12, and 13 have been added as subgeneric and as species of Claim 1 to cover more

specifically the now preferred class of this invention. Support for the added new species claims may be found on pages 10 and 12 of the specification.

Respectfully submitted.

FRANK N. HOUGHTON,
*Agent for Applicant.*

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE

WASHINGTON 25, D. C.

Div. 6, Room 7616. Paper No. 3

Address only: The Commissioner of Patents,-and not any official by name.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the examiner in charge of this application. Casper W. Ooms, Commissioner of Patents.

Applicant: Frederick C. Bersworth. Ser. No. 735,-018. Filed Mar. 15, 1947. For Alkylene Polyamine Derivatives.

FRANK N. HOUGHTON,
*35 East St., Elmwood, Mass.*

This application has been examined.

References cited:

Munz—2,130,505, Sept. 20, 1938, 260–534.

Moore—2,293,034, Aug. 18, 1942 260–482.

Claims 1, 2, 3, 4, 6, 10, and 12 are rejected as being alternative claims in the use of the word "or".

Claims 2, 3, 4, 10, and 12 are rejected as being "Markush" claims of limited scope under the broader "Markush" claim 1 which includes these.

Claim 1 is rejected as lacking invention over each of Munz and Moore. Munz discloses a compound

which differs only in that "A" is a lower homologue of that claimed. Moore discloses a compound which differs only in that it is a derivative of a mono-amine. Had he employed a diamine in the same reactions the claim would have been fully met. These differences do not constitute invention.

Claims 6, 11, and 13 are directed to more than a single species, viz:

1) a glycol tertiary amine
2) a hydroxyl alkali salt carboxylate tertiary amine
3) a hydroxyl ether tertiary amine.

Since no claim generic to these species has been found allowable, election of one of these species is required.

Applicant is advised that his response to be complete under Order 3527 must include an election of species, or a statement that in his opinion all claims are in fact readable on a designated species, which applicant must identify, and that such statement will be construed as an election under the Order.

H. SUBLE,
*Examiner.*

In the United States Patent Office

Div. 6, Room 7616, Elmwood, Massachusetts

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial Number 735,018. Filed March 15, 1947. For Alkylene Polyamine Derivatives.

Hon. COMMISSIONER OF PATENTS,
*Washington, D. C.*

SIR: In response to the Examiner's action of August 4, 1947, please amend the above-entitled application as follows:

Claim 1, seventh line after the formula, cancel "alkyl or hydrogen" and insert—a member of the group consisting of alkyl and hydrogen—.

Claim 2, cancel line 2 and insert — $-CH_2COOM$ wherein M is an alkali metal.

Claim 10 sixth line after the formula, cancel "–OR, wherein R is alkyl or hydrogen" and insert— –OH,–.

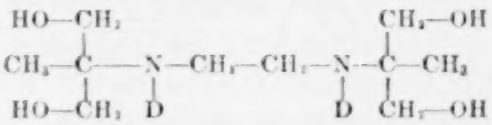
Claim 12 sixth line after formula, cancel "–OR 'OR wherein R is alkyl or hydrogen" and insert– –OR'OH—.

Cancel Claims 3, 4, and 6.

Please add the following claims:

—14. A compound according to claim 1 wherein D is the radical $-CH_2COOH$.

15. A compound having the formula



wherein D is a member of the group consisting of $-CH_2COOH$, $-CH_2CH_2COOH$, and their alkali metal salts.

16. The compound according to claim 15, wherein D is $-CH_2COONa$.—

## Remarks

Claims 1, 2, 10, and 12 have been amended to eliminate any ambiguous use of the word "or"; claims 3, 4, and 6 also objected to on the same ground have been cancelled. Claims 2, 10, and 12 have been amended and are now believed free from objection on the ground of being "Markush" claims of limited scope; claims 3, and 4 have been cancelled.

New claim 14 has been added to supplement claim 2 as amended. New claims 15 and 16 have been added to replace cancelled claim 6.

Claim 1 is rejected as lacking invention over each of Munz and Moore.

Claim 1 is rejected on Munz on the ground that Munz discloses a lower homologue of a species of claim 1.

Applicant is quite familiar with the compound disclosed by Munz and, because of its limitations, was driven to find compounds overcoming those limitations. One of the principal useful characteristics of the compound disclosed by Munz is the power of that compound to sequester polyvalent metal ions. It is, however, incompatible with soap in more than very small amounts; in amounts so small, in fact, that it is useless as an addition to soap for the purpose of softening hard water. The compounds of the present invention, and as defined by claim 1, are compatible with soap and yet have polyvalent metal sequestering power. Moreover applicant's compounds are themselves detergents while that of Munz is not.

Applicant has more than made an analogue of the compound disclosed by Munz—he has made a compound which differs in chemical composition and in action. Munz does not teach nor indicate how the advantages of his compound may be retained and yet overcome its limitations.

Claim 1 was also rejected on Moore for the reason that Moore's compounds are derived from a monoamine. Moore falls far short of disclosing the limitations imposed upon the compounds disclosed and claimed by applicant. One essential characteristic of applicant's compounds is that they must be derived from a polyamine, but more than that they must be derived from a polyamine, the nitrogen atoms of which are separated by "a positive whole number from 2 to 3". If this condition is not met, seemingly similar compounds are of little or no value for applicant's

presently contemplated uses. It is submitted that it is no valid answer to say, "*If* Moore had used a diamine for his starting material instead of a monoamine, *and* if he had selected the proper diamine, *and* if he had made a compound containing, 'a member of the group consisting of $CH_2COOH$, $-CH_2CH_2COOH$, and their alkali metal salts' on each of the nitrogen atoms, *and* if in addition he had insured having the proper chain length and other characteristics of the remaining radicals, he would have applicant's compound." The full equivalency of Moore to applicant's claim 1 is not apparent.

The Examiner has required election of species. Accordingly applicant elects the glycol tertiary amine—now claims 15 and 16. It is requested that the non-elected species be retained in the case pending determination of the generic claims.

The claims now standing in the case, as amended, are believed to be allowable. Allowance is therefore requested.

Respectfully,

FRANK N. HOUGHTON,
*Agent for Applicant.*

In the United States Patent Office

Applicant: Frederick C. Bersworth. Invention: Alkylene Polyamine Derivatives. Examiner's Room 7616, Division 6. Ser. No. 735,018. Filed: Mar. 15, 1947.

NEW POWER OF ATTORNEY

Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: In the above identified application I hereby revoke all previous powers of attorney and appoint

Malcolm N. Rich (Reg. No. 13,142), of 11 Park Place, New York City 7, New York, as my attorney with full power to prosecute said application, to make alterations and amendments therein, to receive the patent, and to transact all business in the Patent Office connected therewith.

Signed at Framingham, Mass., this 17th day of May 1948.

Frederick C. Bersworth,
FREDERICK C. BERSWORTH,
*Applicant.*

MNR:W.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE

WASHINGTON, *June 3, 1948.*

MALCOLM N. RICH,
*11 Park Place,*
*New York 7, N. Y.*

Applicant, Frederick C. Bersworth. Serial No. 735,018. Filed March 15, 1947. For Alkylene Polyamine Derivatives.

In this case your power of attorney has been accepted.

Very truly yours,
LAWRENCE C. KINGSLAND,
*Commissioner of Patents.*

Revoking power of attorney to Frank N. Houghton, 35 East St., Elmwood, Mass.

405368—56——23

Paper No. 7

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Address only the Commissioner of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the examiner in charge of this application. Lawrence C. Kingsland, Commissioner of Patents.

Division: 6—Room 7616 HORNes. Applicant: Frederick C. Bersworth. Ser. No. 735,018. Filed March 15, 1947. For Alkylene Polymine Derivatives. MALCOLM N. RICH,

*11 Park Place New York 7, New York.*

Responsive to amendment of November 5, 1947.

Additional reference:

Munz—2,240,957, May 6, 1941, 252–117.

Claims 1, 2, 10, 12, and 14 are rejected as being alternative in defining "m". (*Ex parte Piggott*, Patent No. 2,091,105.)

Claims 2, 10, 12, 14, 15, and 16 are rejected as being structural formula type claims of diminished scope. The principles of Markush practice apply and in the absence of inventiveness over the broader structure such claims are improper. (*Ex parte Dahlen*, 1934 CD 9 and *Ex parte Cook*, 76 USPQ 196.)

In view of the election of the species of claims 15 and 16, claims drawn to either free acids or any product other than N,N' di-sodium carboxymethyl,N,N' di-dihydroxy (t) butyl ethylene diamine are considered unelected and claims 11, 13, and 14 are correspondingly rejected.

Claims 1, 2, and 14 are rejected as lacking invention over Munz, previously cited. The tetra-acetic acid compound is a lower homologue of the claimed products and the newly cited patent discloses its properties as a sequestering agent and a soap additive. In the absence of a verified showing contrariwise, the properties being similar and the structures being homologues applicant's products are uninventive. (*In re Hass*, 563, 563 OG 576, 579.)

This application should be prepared for final action. No further amendments that do not place the case in condition for allowance or better form for appeal will be entered after final action unless accompanied by a proper showing of good and sufficient reasons why they were not earlier presented. Rule 68 and Commissioner's Notice of April 26, 1948.

H. SUBLE,
*Examiner.*

In the United States Patent Office

Applicant: Frederick C. Bersworth. Examiner's Room 7616. Division number six. Serial No. 735,018. Filed: March 15, 1947. Invention: Alkylene Polyamine Derivaties.

*Amendment (3)*

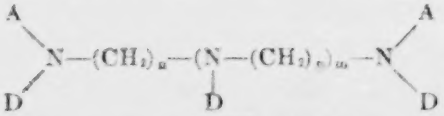
HON. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: In response to the Official Action of September 30, 1948, please amend the above application as follows:

*Claims*

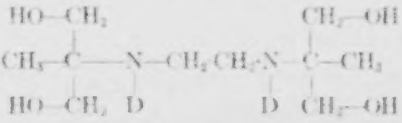
Erase all claims of record and substitute therefor the following claims:

1. A compound having the formula



wherein $n$ is an integer selected from the group consisting of 2 and 3; $m$ has a value equal to one of the group consisting of zero, the numeral 1, and any multiple thereof; D is a member of the group consisting of $-CH_2COOH$, $-CH_2 \cdot CH_2 \cdot COOH$, and their alkali metal salts; A is an alkyl group containing at least seven carbon atoms and having not more than two hydrogen atoms thereof replaced by a member of the group consisting of –COOR, –COOM, –SO H, –SO M, –OR, and –OR′OR wherein R is one of the group consisting of alkyl and hydrogen, M is an alkali metal, and R′ is an alkylene group, each said replacing groups being spaced from the N atoms of the compound by at least two carbon atoms of the hydrocarbon group containing the same.

18. A compound having the formula:



wherein D is a member of the group consisting of $CH_2 \cdot COOH$, $-CH_2 \cdot CH_2 \cdot COOH$, and their alkali metal salts.

19. The compound of claim 18, wherein D is $-CH_2 \cdot COONa$.

20. The compound of N, N′ di-sodium carboxymethyl, N, N′ dihydroxy (t) butyl ethylene diamine.

## Remarks

The above amendment is made for the purpose of simplifying the issue of patentable novelty involved

in the present application by limiting the number of claims to genus and three (3) species.

New genus claim 17, is substantially identical to original claim 1; species claims 18 and 19 are substantially identical to erased elected claims 15 and 16; and species claim 20 defines the elected species chemically in accordance with the definition given by the Examiner in paragraph 3 of the Official Action of September 30, 1948.

As now worded these claims are all believed allowable to the applicant over the art of record, particularly Munz, for the primary reason that the defined compounds *are not homologues* of the compounds disclosed by Munz, as stated by the Examiner.

The compounds of the present invention are derivatives of ethylene diamine and its homologues and differ from those described by Munz in that only two of the amino hydrogens (one each in each amino nitrogen group) have been displaced by the carboxylic acid groups D. The other two amino hydrogens have been displaced by the hydrocarbon A groups. Accordingly, we do not have a homologue or a derivative of ethylene diamine tetra-acetic acid which is the closest carboxylic amino acid disclosed by Munz.

In view thereof, it is believed self-evident that it is not at all obvious that the products of the instant invention should have sequestering powers or should be compatible with soap compounds, as the mechanism involved in sequestering is quite different from that of the compounds disclosed by Munz and an unexpected new result, per se. Moreover, such compounds have soap-like properties and act as *detergents* which also is an unexpected new result.

In consideration of these differences (chemical and physical) between the compounds of Munz and the present compound it is submitted that the Examiner

is in error on the viewpoint expressed in the last paragraph on page 1 of the Action and that this ground for rejection should be withdrawn.

A favorable action with prompt allowance of the new genus and special claims 17 to 20, inclusive, is respectfully requested.

Respectfully submitted,

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His Attorney.*

MARCH 18, 1949.

MNR:W.

POL–90 Paper No. 9

DEPARTMENT OF COMMERCE

UNITED STATES PATENT OFFICE

Washington

Address only the Commissioner of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing and name of the applicant.

Please find below a communication from the EXAMINER in charge of this application. Laurence C. Kingsland, Commissioner of Patents.

Division: 6—Room 7616. Applicant: Frederick C. Bersworth. Ser. No. 735,018. Filed March 15, 1947. For Alkylene Polyamine Derivatives.

MALCOLM N. RICH,

*11 Park Place, New York 7, N. Y.*

Responsive to amendment filed March 19, 1949.

Claims 17 to 20 appear in this case.

The specification (page 7, lines 6–10) should be corrected by inserting a $CH_2$ between the carbonyl radical and the nitrogen atom. It is evident that applicant intends the structure, as suggested, rather than the carbamate structure, as shown. In the absence of such a correction, claims 19 or 20 may not be allowed, since the only disclosure of the compounds, as claimed, consists of the compound, as corrected, on page 7 of the specification.

Claim 17 is *finally* rejected as being unpatentable over Munz, previously cited. Munz shows an ethylene diamine with 2 acetic acid radicals attached to each nitrogen atom, which is a lower homologue of the compound as claimed. A homologue of a known compound is not patentable in the absence of a verified showing of unexpected properties. In re Hass 563 O. G. 576, 579. The Arguments of applicant are noted and it is pointed out that the only difference between the compound of Munz and the compound as claimed is the presence of 3 additional $CH_2$ groups in one of the groups attached to each nitrogen atom, in the compound as claimed. It is pointed out that the A group of the compound as claimed contains the group $-CH_2CH_2CH_2CH_2COOR$, while the D group contains the acetic acid group. One of the two acetic acid groups on each nitrogen atoms of Munz may be considered as being D. The other acetic acid group ($-CH_2COOR$) may be considered as A. Therefore, while A of the compound, as claimed, is $-(CH_2)_4COOR$, A of Munz is $-(CH_2)COOR$, resulting in a difference between the compounds of only $(CH_2)_3$. Homologues are compounds differing in structure by $(CH_2)_n$. Therefore, the compound as claimed is a homologue of the compound cited as a reference. It is also pointed out by Munz (2,240,957) (page 2, col-

umn 2, lines 55–75) that the compound of the reference is used alone in aqueous solution for deterging and washing. Therefore it has a soap-like and detergent quality. Furthermore, Munz's compound is used in conjunction with soap and therefore it is implied that the two are compatible. Munz (page 1, column 1, lines 44 to 54) teaches that the compounds of the reference have sequestering powers. The compound of the reference is used to make hard water soft, as is the compound as claimed. Applicant insists, by remarks in the amendments, that the compounds as claimed have unexpected properties over the compound disclosed, but has offered no verified showing of such unexpected properties. It is submitted that such remarks are inadequate to establish the unexpected properties required by In re Hass.

Claim 18 is finally rejected as being a structural formula type claim of diminished scope. The principles of Markush practice apply and in the absence of inventiveness over the broader structure, such claims are improper. (*Ex parte Dahlen* 1934 C. D. 9 and *Ex parte Cook* 76 U. S. P. Q. 196.) Markush groups of diminishing scope are improper no matter how few the number of claims in the case. (*Ex parte Kline* 25 J. P. O. S. 767.) Since the Markush type claim of claim 2 does not have properties not shared by the true genus as a whole and which patentably distinguish from the true genus as a whole, it is improper. (*In re Thompson et al.* 1946 C. D. 280.)

Claims 19 and 20 are objected to as containing identical subject matter and being of identical scope, but otherwise appear to be allowable. One of these two claims should be selected for purposes of allowance.

This section is made *FINAL*. With reference to any further action that may be taken in this case attention is directed to new Rules 116 and 191–195 now in force.

WILLIAM G. WILES,
*Acting Examiner.*

In the United States Patent Office

Applicant: Frederick C. Berswerth. Division: #6. Room: #7616. Serial No. 735,018. Filed: Mar. 15, 1947. Invention: Alkylene Polyamine Derivatives.

*Amendment (4)*

HON. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: Please amend the above application as follows:

SPECIFICATION

Page 4, in the formula between lines 4 and 3 (from bottom) change the $(CH)_2$ group to—$(CH_2)_2$—.

Page 7, in the formula between lines 5 and 6 insert—$CH_2$—between ~~the carbonyl~~ radical and the nitrogen atom on each side of the —$CH_2CH_2$—group.

Page 8, in the formula between lines 2 and 3 (from bottom) change the $(CH_2)$ group in the side chains from each CH group attached to the nitrogen atom to —$(CH_2)$—.

CLAIMS

Claim 17, line 8, erase "two" insert—seven—; line 11, erase "$(CH_2)_2$" (each occurrence); line 12, before "alkyl" insert—consisting of—.

Claim 18, lines 2 to 4, erase "a member of ~~_____~~ metal salts" insert—acetic acid—.

Erase claim 19.

## Remarks

The above amendment is made for the purposes of allowance or appeal.

The amendments to the specification in the formulas on pages 4, 7 and 8 are to correct obvious errors.

Claim 19 has been erased in an election between identical claims.

Claim 17 has been limited as to the "A" group so as to define the compound completely away from the Munz patent and thereby to avoid the Examiner's grounds for rejecting the claim on this patent.

Claim 18 has been amended to limit same to a species "D" group.

The compound of claim 17 as now defined generically is not a homologue of the compound disclosed by Munz because the Munz compounds require these "A" groups to be acetic acid groups not long chain fatty acid groups. The inventive thought of *substituting such long chain fatty acid groups for an amino hydrogen in "A" position in ethylene diamine is clearly lacking in Munz.*

These groups change profoundly the chemical properties of the compound, *imparting* soap-like properties thereto and surface acting properties without losing chelating properties normally present in the tetra-acetic acid compound. The Munz compounds are *only* chelating compounds which are useful *in association* with soap compounds. They are not detergents, per se, nor are they surface acting agents, per se.

The above amendment is made pursuant with an agreement reached with the Examiner in an informal interview at which the above essential points of difference between applicant's compound and the compound of the Munz patent were explained and argued. The Examiner agreed that by limiting this claim 17

to "A" groups consisting of long chain fatty acids the claim would be allowable.

As the above amendments were agreed on at the interview the claims are believed allowable on inspection and such action is requested promptly to avoid the necessity of filing an appeal.

Respectfully submitted,

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His Attorney.*

MNR: W.
February 28, 1950.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE

Washington

POL–90 Paper No. 11

Address only the Commission of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the EXAMINER in charge of this application. John A. Marzall, Commissioner of Patents.

Division: 6–Room 7616. Applicant: F. C. Bersworth. Ser. No. 735,018. Filed Mar. 15, 1947. For Alkylene Polyamine Derivatives.

MALCOLM N. RICH,
*11 Park Place, New York 7, N. Y.*

Responsive to the amendment of March 1, 1950.

The above identified amendment has not been entered since it does not place the application in condition for allowance. The amendment will be entered for purposes of appeal, if applicant desires. The

claims amended as proposed will be rejected for the following reasons.

Claims 17, 18 and 20 are drawn to three independent species; i. e., (1) A containing at least seven carbon atoms (2) A containing 4 carbon atoms and D being acetic acid (3) A containing 4 carbon atoms and D being a sodium acetate radical.

Since there is no claim generic to each of these three species, the case is not in condition for allowance. In the event that an allowable generic claim be submitted, it is felt that, as at present advised, the claims contain allowable subject matter.

H. SUBLE, *Examiner.*

In the United States Patent Office

Applicant: Frederick C. Bersworth. Division: #6. Room: #7616. Serial No. 735,018. Filed: Mar. 15, 1947. Invention: Alkylene Polyamine Derivatives.

*Amendment (5)*

HON. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: Please amend the above identified application as follows:

CLAIMS

Claim 17, line 7, erase "hydrocarbon" and insert—alkyl—.

Erase claims 18 and 20 and insert the following claims 21 and 22:

21. The compound of N.N' di-octyl ethylene diamine N.N' di-acetic acid.

"22. The compound N,N' di-9-hendecenoic ethylene diamine N,N' di-acetic acid and its salts and derivatives."

## Remarks

Claim 17 has been amended to limit same to alkyl group containing at least 7 carbon atoms.

Claims 18 and 20 indicated as being allowable are erased as the subjects matter thereof are covered in the claims of copending application Serial No. 39,377 filed July 17, 1948. New species claims 21 and 22 are inserted for the purpose of allowance or appeal along with allowable generic claim 17.

Referring to claim 21, the compound defined by this claim was claimed originally sub-generically in original claim 3 and forms the subject matter of example 1 on page 4 of the specification.

Referring to claim 22, this compound was claimed originally in original claim 5 and forms the subject matter of the example on page 8 of the specification.

Each of the compounds of claims 21 and 22 are broadly included within the scope of allowable claim 17 and are species claims to claim 17. Claim 17 being indicated as being allowable there seems to be no reason why these species claims also cannot be allowed on inspection. As these compounds were claimed originally and are the subject matter of specific examples no new search is believed necessary in view of the patentability of generic claim 17 before the Examiner can act on these claims on inspection.

A prompt allowance of this application on inspection is respectfully solicited.

Respectfully submitted.

Frederick C. Bersworth,

By Malcolm N. Rich,

*His Attorney.*

March 7, 1950.

MNR:hgn.

In the United States Patent Office

Applicant: Frederick C. Bersworth. Division # 6. Room: # 7616. Ser. No. 735,018. Filed: Mar. 15, 1947. Invention: Alkylene Polyamine Derivatives.

*Supplemental Amendment* (5)

Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: Please amend the above application as follows:

SPECIFICATION

Page 4, line 25, erase "has" insert—is the sodium salt of N,N′ di-octyl ethylene diamine N,N′ diacetic acid having

Page 8, line 22, erase "$CH_3$—COOH" insert—

$$CH_3—CH=CH-(CH_2)_7—COOH$$

—; line 23, erase "the formula—as follows" insert—the product is the di-methyl ester of ethylene diamine N,N′ di (sodium acetate) N,N′ di (9-ethyl nonylic acid) having the folowing formula:

CLAIMS

Claim 21, lines 2 and 3, erase "and its salts and derivatives".

Erase claim 22 and rewrite same as new claim 23 reading as follows:

3. The compound ethylene diamine N,N′ di-acetic acid N,N′ di (9-ethyl nonylic acid).

REMARKS

The above amendment to the specification and claims is made in accordance with the request of the

Examiner to clarify the specification and to overcome objections as to the form of the claims 21 and 22.

As the period of response expires on March 12, 1950, an appeal is filed herewith on claims 17, 22 and 23, to save the application from abandonment.

Early allowance of this application is solicited.

Respectfully submitted.

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His Attorney.*

MARCH 10, 1950.
MNR:W.

In the United States Patent Office

Applicant: Frederick C. Bersworth. Room: #7616. Division: #6. Serial No. 735,018. Filed: Mar. 15, 1947. Invention: Alkylene Polyamine Derivatives.

APPEAL TO THE BOARD OF APPEALS

Hon. COMMISSIONER OF PATENTS,
*Washington, 25, D. C.*

SIR: I hereby appeal to the Board of Appeals from the decision of the Principal Examiner in finally rejecting claims 17, 21 and 23, of the above identified application.

The following are the points of the decision on which the appeal is taken:

1. The Principal Examiner erred in finally rejecting the claims upon the references of record.

2. The Principal Examiner erred in finally rejecting the claims for the reasons of record.

3. The Principal Examiner erred in not allowing any or all of these claims.

The appeal fee of $15.00 is enclosed herewith. An oral hearing is requested at the convenience of the Board.

Respectfully,

FREDERICK C. BERSWORTH,
By MALCOLM N. RICH,
*His Attorney.*

MARCH 10, 1950.
MNR:W.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

POL–37 PAPER No. 15

Address only
The Commissioner of Patents
Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the Examiner in charge of this application. John A. Marzall, Commissioner of Patents.

Division: 6, Room 7616. Applicant: Frederick C. Bersworth. Ser. No. 735,018. Filed March 15, 1947. For Alkylene Polyamine Derivatives.

MALCOLM N. RICH, 11 Park Place, New York 7, N. Y.

The changes hereinafter specified have been made in the application identified above in accordance with the provisions of Order Number 2308 of March 12, 1917, which reads in part as follows:

> Obvious informalities in the application may be corrected by the examiner, but said correction must be in the form of an amendment, ap-

proved by the Principal Examiner in writing, placed in the file, and made a part of the record. The changes specified in the amendment will be entered by the clerk in the regular way.

If these changes are not satisfactory to the applicant, appropriate amendment may be proposed under the provision of Rule 312, provided the printing of the specification has not begun at the time such amendment is ready for entry. To insure the consideration of such amendments for inclusion they should be submitted on or before the date of remitting the final fee since the printing of the specification begins a few days thereafter.

The application has been amended as follows:

Page 8, in the formula between lines 2 and 3 (from bottom) "$(CH_2)_x$" has been changed to $-CH_2-$.

Since applicant obviously intends to disclose and claim the 9-ethyl nonylic acid radical attached to both nitrogen atoms, Examiner has amended the amendment of March 2, 1950 (paper #10) concerning the formula on page 8, to read as originally presented, since the amendment of March 2, 1950 obviously introduces an error.

H. SURLE,
*Examiner.*

405368—56——24

DEPARTMENT OF COMMERCE

UNITED STATES PATENT OFFICE

Washington

POL–85

Address only The Commissioner of Patents, Washington 25, D. C.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

MAY FOURTH, 1950.

MALCOLM N. RICH, *11 Park Place, New York 7, N. Y.*

NOTICE OF ALLOWANCE

The application for patent identified below has been examined and found allowable for issuance of Letters Patent.

Applicant: Frederick C. Bersworth.

Title of Invention or Improvement: Alkylene Polyamine Derivatives.

Serial No. 735,018.

Filing date, March 15, 1947.

Number claims allowed, 3.

With the allowance of the application the final fee becomes due. This fee is thirty dollars ($30) plus one dollar ($1) for each claim allowed in excess of twenty (20) and must be paid within 6 months from the date of this notice. Failure to remit the final fee will result in the patent being withheld from issue. In remitting this fee identify the application to which it applies by giving the followng informaton: Name of inventor, title of invention, serial number, date of filing, date of allowance, and if assigned, the names of the assignees. The final fee will not be received from anyone other than the applicant, his assignee or

attorney, or a party in interest as shown by the records of the Patent Office.

If it is desired to have the patent issued to an assignee or assignees an assignment containing a request to that effect, together with the fee for recording the same, must be filed in this Office on or before the date of payment of the final fee.

The patent will be issued and forwarded approximately 4 weeks after receipt of the final fee.

By direction of the Commissioner.

H. SURLE,
*Examiner.*

REFERENCES CITED

The following references are of record in the patented file of this patent:

UNITED STATES PATENTS

| *Name* | *Number* | *Date* |
|---|---|---|
| Munz | 2,130,505 | Sept. 20, 1938 |
| Munz | 2,240,957 | May 6, 1941 |
| Moore | 2,293,034 | Aug. 18, 1942 |

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Paper No. 17

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Address only: The Commissioner of Patents, Washington 25, D. C.

Please find below a communication from the examiner in charge of this application. John A. Marzall, Commissioner of Patents.

Division: 6. Applicant: Frederick C. Bersworth. Ser. No. 735,018. Filed March 15, 1947. For Alkylene Polyamine Derivatives.

MALCOLM N. RICH,
*11 Park Place, New York 7, New York.*

The changes hereinafter specified have been made in the application identified above in accordance with the provisions of Order Number 3141 of February 4, 1930, (Manual of Patent Examining Procedure 1302.04), which reads in part as follows:

> Other obvious informalities in the application may be corrected by the examiner, but such corrections must be in the form of an amendment, approved and signed by the Principal Examiner, placed in the file, and made a part of the record. A copy of the amendment is sent to the applicant. The changes specified in the amendment will be entered by the clerk in the regular way.

If these changes are not satisfactory to the applicant, appropriate amendment may be proposed under the provision of Rule 312, provided the printing of the specification has not begun at the time such amendment is ready for entry. To insure the consideration of such amendments for inclusion they should be submitted on or before the date of remitting the final fee since the printing of the specification begins a few days thereafter.

The application has been amended as follows:

In the specification page 13, last line after "1946" the following has been inserted: —, which issued as Patent No. 2,524,218 on October 3, 1950.

H. SURLE,
*Examiner.*

FINAL FEE PAID TO THE COMMISSIONER OF PATENTS

(Be careful to give correct Serial No.)

NOVEMBER 2, 1950.

Serial No. 735,018

Inventor: Frederick C. Bersworth. Patent to be issued to Frederick C. Bersworth. Name of invention, as allowed: Alkylene Polyamine Derivatives.

Date of payment: . Fee: $30.00.

Date of filing: Mar. 15, 1947. Date of circular of allowance: May 4, 1950.

The Commissioner of Patents will please apply the accompanying fee as indicated above.

MALCOLM N. RICH,
*Attorney.*

Send patent to Malcolm N. Rich, 11 Park Place, New York 7, N. Y.

Final fees will not be received from other than the applicant, his assignee or attorney, or a party in interest as shown by the records of the Patent Office, nor will they be applied in pending applications.

NUMBER (Series of 1935)
735018

PATENT NO.
1947 DATED 2532391

DIV. — ✓

(EX'R'S BOOK) 43-48-59

Name FREDERICK C. BERSWORTH

of VEROMA

State of NEW JERSEY

Invention ALKYLENE POLYAMINE DERIVATIVES

ORIGINAL

APPLICATION FILED COMPLETE MAR 15, 1947

PARTS OF APPLICATION FILED

Petition, Specification, Oath, First Fee $30 ~~Sheets Drawings~~, MAR 15, 1947

Examined and passed for Issue MAY 4 1950, 194 ... Exr. Div. 6

Notice of Allowance ... Mail 4 ..., 194

Final Fee $30 Nov 3/50 ..., 194

Attorney ~~FRANK N. HOUGHTON 35 EAST ST ELMWOOD MASS.~~

Associate Attorney ...

No. of Claims Allowed 3 Print Claims 1 in O. G. Class 260-439.5

Title as allowed Alkylene ... Derivatives

PAPER NO. 2

# 1947

MAR 10 1947

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*Plaintiffs' Exhibit No. 17*

I claim:

1. The process of preparing aliphatic polycarboxylic amino acids which comprises reacting an aliphatic amine having at least one replaceable hydrogen atom attached directly to an amino nitrogen atom, with an alkali metal cyanide and a formaldehyde-yielding substance under conditions sufficiently alkaline that there is substantially no hydrolysis of the said cyanide, while maintaining the amount of said free cyanide present in the reaction mixture at any one time at not more than about one quarter of the amount necessary to react completely with the original amine, and maintaining the amount of free formaldehyde-yielding substance present at any one time at less than one mol thereof per mol of said free cyanide present until substantial completion of the reaction of said amine has been effected, and removing ammonia from the reaction mixture so that substantially no free ammonia is present therein.

2. The process according to claim 1 wherein the reaction is carried out at a temperature of between about 60° C. and about 80° C. and at a pressure such that the reaction mixture is substantially at the boiling point at said temperature, until the major part of the reaction has been completed, and the temperature and pressure are then gradually raised while maintaining substantially boiling conditions, until completion of the reaction.

3. The process of preparing aliphatic polycarboxylic amino acids which comprises reacting an aliphatic amine having at least one replaceable hydrogen atom attached directly to an amino nitrogen atom, with an alkali metal cyanide and a formaldehyde-yielding substance under conditions sufficiently alkaline that there is substantially no hydrolysis of the

said cyanide while maintaining the amount of said free cyanide present in the reaction mixture at any one time at not more than about one quarter of the amount necessary to react completely with the original amine, and maintaining the amount of free formaldehyde-yielding substance present at any one time at less than one mol thereof per mol of said free cyanide present until substantial completion of the reaction of said amine has been effected, and removing ammonia from the reaction mixture so that substantially no free ammonia is present therein at any time during the reaction, and finally, after all of the cyanide has been added, adding a slight excess of formaldehyde-yielding substance over that necessary to react with any unreacted cyanide.

4. The process according to claim 1 wherein the formaldehyde-yielding substance is formaldehyde.

5. The process according to claim 1 wherein the amine is a primary aliphatic amine.

6. The process according to claim 1 wherein the amine is a primary aliphatic diamine.

7. The process of preparing the sodium salts of ethylene diamine tetraacetic acid which comprises reacting ethylene diamine with sodium cyanide and formaldehyde in the presence of sufficient sodium hydroxide to bring the pH to at least about 9 while maintaining a temperature of between about 60° C. and about 80° C. and a pressure such that the reaction mixture is substantially at the boiling point at said temperature, and maintaining the amount of free cyanide present in the reaction mixture at any one time at not more than one quarter of the amount necessary to react completely with the ethylene diamine, and maintaining the amount of free formaldehyde present in the reaction mixture at any one time at about five per cent less than the amount of

sodium cyanide present, which five per cent is based on the total quantity of sodium cyanide to be reacted, until at least about 95 per cent of the theoretically required four mols have been added, adding sufficient additional formaldehyde substantially completely to react with the unreacted sodium cyanide, increasing the pressure and heating the solution to boiling at a temperature at least about 110–120° C., and removing ammonia from the reaction mixture so that substantially no free ammonia is present therein at any time.

8. The process of preparing acid salts of aliphatic polycarboxylic amino acids which comprises reacting an aliphatic diamine having two replaceable hydrogen atoms attached directly to each amino nitrogen atom, with an alkali metal cyanide and a formaldehyde-yielding substance under conditions sufficiently alkaline that there is substantially no hydrolysis of the said cyanide, while maintaining the amount of said free cyanide present in the reaction mixture at not more than about one quarter of the amount necessary to react completely with the original diamine, and maintaining the amount of free formaldehyde-yielding substance present at less than one mol thereof per mol of said free cyanide present until substantial completion of the reaction of said diamine has been effected, removing ammonia from the reaction mixture so that substantially no free ammonia is present therein, and then precipitating the resulting tetraacetic acid by lowering the pH of the reaction mixture to about .75 to 2, adding the precipitated acid to about four times the weight of the sodium salt of said acid, in the presence of water, and recovering the diacetic acid-disodium salt crystallizing out therefrom on cooling.

9. The process of preparing the mono-sodium salt of ethylene diamine tetraacetic acid which comprises

reacting ethylene diamine with sodium cyanide and formaldehyde in the presence of sufficient sodium hydroxide to bring the pH to at least about 9, while maintaining a temperature of between about 60° C. and about 80° C. and a pressure such that the reaction mixture is substantially at the boiling point at said temperature, and maintaining the amount of free cyanide present in the reaction mixture at any one time at not more than one quarter of the amount necessary to react completely with the ethylene diamine, and maintaining the amount of free formaldehyde present in the reaction mixture at any one time at about five per cent less than the amount of sodium cyanide present, which five per cent is based on the total quantity of sodium cyanide to be reacted, until at least about 95 per cent of the theoretically required amount of formaldehyde has been added, adding sufficient additional formaldehyde substantially completely to react with the unreacted sodium cyanide, increasing the pressure and heating the solution to boiling at a temperature at least about 110–120° C., removing ammonia from the reaction mixture so that substantially no free ammonia is present therein at any time, acidifying the resulting product to a pH value between about .75 and about 2 and precipitating ethylene diamine tetraacetic acid, and then mixing said precipitated acid with a substantially saturated solution of the dialkali metal salt of aminocarboxylic tetraacetic acid in equimolecular proportions, heating the solution until the acid dissolves, and recovering the resulting monoalkali metal salt by evaporation of the water.

10. The process according to claim 1 wherein the amine is diethylene triamine.

11. The process according to claim 1 wherein the amine is n-butylamine.

12. The di-alkali-metal salt of ethylene diamine tetraacetic acid.

13. The mono-alkali-metal salt of ethylene diamine tetraacetic acid.

14. The penta-alkali-metal salt of a dialkylene triamine pentaacetic acid.

15. A dialkylene triamine pentaacetic acid.

16. A polyalkylene polyamine containing both amino and imino groups, and having each of its amino and imino hydrogen atoms replaced by the radical $-CH_2COONa$.

17. A polyalkylene polyamine containing both amino and imino groups, and having each of its amino and imino hydrogen atoms replaced by the radical $-CH_2COOH$.

18. The process and/or product and/or composition having any one or more of the novel characteristics herein shown and/or described.

Signed by me at Newark, New Jersey this 16th day of June, 1943.

(Inventor's Signature) FREDERICK C. BERSWORTH.

OATH

STATE OF NEW JERSEY,

*County of Essex, ss:*

Frederick C. Bersworth, the above-named petitioner, being sworn, deposes and says that he is a citizen of the United States of America and resident of Verona, New Jersey, that he verily believes himself to be the original, first, and sole inventor of the inventions in Aliphatic Polycarboxylic Amino Acids and Process of Making Them, described and claimed in the annexed specifications; that he does not know and does not believe that the same were ever known or used before his invention or discovery thereof, or patented or described in any printed publication in

any country before his invention or discovery thereof, "or in public use or on sale in the United States for more than one year prior to application Serial No. 400,967, filed July 3, 1941, of which this is in part a continuation as to the subject matter therein, nor more than one year prior to this application as to the additional subject matter herein;" that said inventions have not been patented in any country foreign to the United States on an application filed by him or his legal representatives or assigns more than twelve months prior to this application; and that no application for patent on said inventions has been filed by him or his representatives or assigns in any country foreign to the United States.

(Inventor's Signature) FREDERICK C. BERSWORTH.

Subscribed and sworn to before me this 6th day of June 1943.

[SEAL] HAROLD O. SPARKS,
*Notary Public.*

A Notary Public for New Jersey. Commission Expires 10/31/43.

In the United States Patent Office

Division 6. Elmwood, Massachusetts. July 15, 1943.

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 491,669. Filed June 21, 1943. Aliphatic Polycarboxylic Amino Acids and Process of Making Them.

Hon. COMMISSIONER OF PATENTS,
*Washington, D. C.*

SIR: Please amend the above-entitled application as follows:

Page 10 line 29 after "Serial No." insert "—491,670—."

Same line, after "filed" insert "—June 21, 1943—."
Respectfully.

FRANK N. HOUGHTON,
*Agent for Applicant.*

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Richmond, Va.

Div. 6, Room 272.

Paper No. 3

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Address only: The Commissioner of Patents, and not any official by name.

Please find below a communication from the examiner in charge of this application.

Conway P. Coe, Commissioner of Patents.

Applicant: Frederick C. Bersworth. Ser. No. 491,669. Filed June 21, 1943. For Aliphatic Polycarboxylic Amino Acids and Process of Making Them.

FRANK N. HOUGHTON,
*35 East St., Elmwood, Mass.*

This application has been examined.

References made of record:

I. G. Farben (Dutch)—46,892, Sept. 16, 1939, 260/534.

I. G. Farben (British)—490,384, Aug. 15, 1938, 260/534.

I. G. Farben (French)—730,760, Aug. 24, 1932, 260/534.

Ulrich et al.—2,168,181, Aug. 1, 1939, 260/534.

Munz—2,130,505, Sept. 20, 1938, 260/534.

The Dutch and British patents cited apparently correspond to German patents #664,499 and

#671,334. The French patent apparently corresponds to British patent #388,874.

Claim 18 is rejected as being non-statutory.

Claims 12 to 17 inclusive are rejected as being fully met by the Dutch or British patents cited above.

Claims 1 to 11 inclusive are rejected as being fully met by the Dutch patent cited above or unpatentable over this patent in view of the French patent cited above. The Dutch patent apparently refers to the process as a method of manufacture of the products. However, the process is disclosed by the French patent. The process of making methylene acid, nitrile or chloride addition products using formaldehyde in the presence of the nitrile or chloride ion is old as disclosed.

No generic claim having been allowed, tentative election is required as follows:

(I) To a di-alkali metal salt (claims 8, 12).

(II) To a mono-alkali metal salt (claims 9, 13).

(III) To a penta-alkali metal salt (claim 14).

(IV) To an acid alkali metal salt (claims 10, 15, 17).

(V) To an acid deriv. of butylamine (claims 5, 11).

Claim 16 is generic to groups I, II and III. Claims 1 to 4, 6 and 7 may be read with either group. Ultimate restriction to three species will be required.

The applicant is advised that his response to be complete under Order 3527 must include an election of species, or a statement that in his opinion all claims are in fact readable on a designated species, which applicant must identify, and that such statement will be construed as an election under the Order.

E. W. GENIESSE,
*Examiner.*

## In the United States Patent Office

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 491,669. Filed June 21, 1943. Aliphatic Polycarboxylic Amino Acids and Process of Making Them. Division 6, Room 272.

ELMWOOD, MASSACHUSETTS,
APRIL 17, 1944.

HON. COMMISSIONER OF PATENTS,
*Washington, D. C.*

SIR: In response to Examiner's action of November 10, 1943, please amend the above-entitled application as follows:

*Claims 16 and 17*, Line 1 of each, after "polyamine" insert—containing both amino and imino groups, and—

Line 2 of each—before "hydrogen" insert—and imino—.

Cancel claims 8, 9, 12, 13 and 18.

### REMARKS

Claims 12 to 17 are rejected as being met by the Dutch or British patents cited. Claims 12 and 13 have been cancelled. The remaining claims 14 through 17 refer to polyalkylene polyamine reaction products, and in each case each of the amino and imino hydrogen atoms have been replaced by a carboxyl radical. Neither the British patent 490,384, nor the Dutch patent 46,892 shows that any of the above compounds have been prepared. The claims to the products, now that claims 16 and 17 have been amended, all are directed toward products in which an imino hydrogen as well as the amino hydrogens have been replaced by carboxyl groups. The cited patents do state that such compounds as polyalkylene polyamines

may be used as reactants but they do not show that any product containing more than four carboxyl groups included in one molecule has been made nor do they show the reaction involving the relatively difficulty replaceable imino hydrogen. In view of the statement made on page 9, line 7 of applicant's specification and in view of the fact that the patents cited are concerned with the chloracetic acid method, there is no showing whatever that reference is being made in the cited patents to compounds in which all the amino and imino hydrogens have been replaced by $-CH_2COOH$ or $-CH_2COONa$ groups. In any case such compounds are not shown by the references nor can they be inferred in view of the difficulty involved in their preparation. It is not obvious chemistry that both amino and imino hydrogens will react by the chloracetic acid method. Claims 14 through 17 are therefore believed allowable.

Claims 1 to 11 are rejected on the Dutch patent alone or the Dutch patent in view of the French patent 730,760. Of these claims, claims 8 and 9 are now cancelled.

The Dutch patent refers to the reaction of chloracetic acid with alkyl diamines and in particular with ethylene diamine or polyalkylene polyamines such as diethylene triamine. Such reactions are now old and are not involved in the process of applicant.

The Dutch patent also is concerned with the reaction of chloracetic acid with the products which are formed by the condensation of polyalkylene polyamines with formaldehyde. Applicant uses no chloracetic acid in his process nor does he follow the same procedures. The Dutch patent also reacts the dimethyl or diethyl esters of iminodiacetic acid with the methyl or ethyl esters of oxalic or malonic acids—

such a procedure is unknown to applicant, and in any case is not within the scope of his claims.

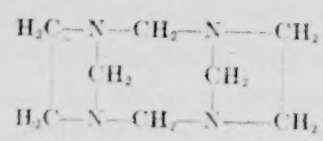
The Dutch patent mentions no reaction with cyanide while applicant's process claims (1 to 7, 10 and 11) are all to cyanide reactions. Therefore the Dutch patent cannot anticipate applicant's process. The formaldehyde reaction of the Dutch patent is an amine coupling reaction in no wise related to the cyanide-formaldehyde reaction of the applicant.

It is therefore submitted that the process claims (1 to 7, 10 and 11) are allowable over the Dutch patent 46,892.

The French patent 730,760 shows that, in general, derivatives of amino carboxylic acids may be obtained by the reaction of HCN or compounds which release it, on the products of the reaction of aldehydes or ketones on primary and secondary amines. In any case the HCN may be introduced as a gas or formed in the reaction chamber by the reaction of an acid on a cyanide salt. Example I of the French patent shows the addition of formaldehyde to the amine. After the reaction has been completed, NaCN is added together with $CO_2$ (so that HCN may be formed). At the completion of this reaction NaOH is added to hydrolyze the nitrile formed.

It is to be pointed out that such a process is altogether different from applicant's and, as a matter of fact, gives particularly poor yields of the final product. In the first place applicant has some of each reactant present at all times and does not perform stepwise reactions as called for in the French patent. In the second place the cited patent reaction involving the cyanide takes place in an acid medium which is the previously known method, and involves the use of a highly toxic and less satisfactory reactant than sodium cyanide. Applicant was careful to avoid ex-

cessive amounts of free formaldehyde in the presence of the amine because it lowered the yield of the final product. According to authorities (Beilstein IV p. 250) (Bischoff B. 31, 3254) a base $C_6H_{12}N_4$ whose structure is thought to be:



is formed by the reaction of formaldehyde with ethylene diamine at ordinary temperatures in aqueous solution.

Appreciation of the necessity for less than total addition of the cyanide and particularly for keeping the concentration of formaldehyde low, which is lacking from the French reference, is an important contribution to the art. The carrying out of the reaction at such a high pH that no HCN is ever present is also a valuable contribution and a sharp differentiation from the French patent.

Still another of applicant's contributions which greatly increases the yield is the continual removal of nitrile, as fast as it forms, by hydrolysis. In this way the concentration of nitrile at any time is substantially zero, thus preventing side reactions and any tendency to reversal of the reaction. This also is a sharp distinction from the French patent, and is clearly recited in the claims.

In Example II of the French patent formaldehyde is reacted with the cyanide, and this reaction product is reacted with the amine. Later the product is hydrolyzed.

As pointed out in applicant's specification on page three beginning with line 9, such a procedure is to be avoided as small yields are obtained due to the formation of glycolates. Applicant's claims definitely avoid

such a process. Example 3 of the French patent involves the use of HCN gas and is of no interest. Examples 4, 5 and 9 also are limited to HCN.

The Examiner has required election as no generic claim has been allowed. It is thought that upon reconsideration the Examiner will find that a generic claim is allowable, but in conformity with his requirement, applicant elects species III as designated by the Examiner. As claims 8, 9, 12, and 13 have been cancelled species I and II need no longer be considered. Species III, IV, and V remain.

Applicant does not understand the basis for and designation of species IV; claim 10 is directed to the use of diethylene triamine according to the process of claim 1—this will result necessarily in a penta-acetic acid alkali metal *salt;* claim 15 is to a penta-acetic *acid;* claim 17 is directed to an acid having at lesat 5 carboxyl groups. Applicant also has some questions as to other statements by the Examiner in regard to the election,—thus, claim 16 is generic to species III but not to species I and II; claim 5 is generic to *all* species and does not come under species V alone, as it refers to amines generally and is not limited to monoamines or to butylamine. If the Examiner is not in agreement with these conclusions, his comments thereon will be appreciated.

The patents to Ulrich and Munz, cited but not applied, are noted, but since they both deal with the chloracetic acid process, they are not believed to be pertinent.

In view of the foregoing remarks, and the amendments presented herewith, it is believed that this application is in condition for allowance. Reconsidera-

tion and allowance are accordingly respectfully solicited.

Respectfully,

FREDERICK C. BERSWORTH,
By FRANK N. HOUGHTON,
*His Agent.*

Div. 6, Room 272 Paper No. 5

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Richmond 20, Virginia

Address only the Commissioner of Patents, and not any official by name.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the examiner in charge of this application. Conway P. Coe, Commissioner of Patents.

Applicant: Frederick C. Bersworth. Ser. No. 491,669. Filed June 21, 1943. For Aliphatic Polycarboxylic Amino Acids and Process of Making Them. FRANK N. HOUGHTON,

*35 East St., Elmwood, Mass.*

Responsive to amendment filed Apr. 19, 1944.

Additional references:

Fick et al. (German)—673,590, Mar. 28, 1939, 260/534.

Polstorff et al., Ber. der Deutschen Chem. Gesell., Vol. 45, pp. 1905–12.

Claims 1 to 7 inclusive, 10 and 11 are rejected as being unpatentable over the Berichte reference cited above in view of either the French patent of record, or the German patent cited above and either the Dutch patent or the British patent of record. The first two references fully set forth the process and

conditions, while the last three patents disclose the reaction applied to polyamines including diethylene triamine and the products. The Polstorff et al. reference indicates that the nitrile group is not hydrolysed in neutral or slightly acid solution. On this basis applicant's functional statement, "under conditions sufficiently alkaline", appears to be too broad and ambiguous. The French patent discloses preparation of the nitrile without hydrolysis. The proportions of reagents seem to be implied by the references, or could be determined by average skill. The Polstorff et al. teaches that the materials will react with amines and ammonia. Furthermore, in view of the breadth of the claims, the references are considered to teach the subject matter.

Claims 14 to 17 are again rejected as being unpatentable over the Dutch or British patents of record or the Fick et al. patent cited above. The patents clearly refer to the reaction between diethylene triamine and chloracetic acid, which would prepare applicant's products, and refers to the acids and products resulting from the reaction. The German patent cited above apparently describes compounds fully meeting claims 16 and 17 and, by implication, claims 14 and 15 in Example 3. The compound is a polyamine and contains imino groups and all free hydrogens on the amino groups are substituted as in applicants compounds. Claims 16 and 17 do not necessarily cover penta-acetic compounds, since some of the amino hydrogens may be substituted by alkyl groups. They are thus ambiguous and too broad.

Claims 14, 15 and 17 are rejected as failing to read on the elected species. There is no claim generic to (A) claims 14 and 16 and (B) claims 15 and 17.

E. W. GENIESSE,
*Examiner.*

In the United States Patent Office

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 491,669. Filed June 21, 1943. Aliphatic Polycarboxylic Amino Acids and Process of Making Them. Division 6, Room 272.

ELMWOOD, MASSACHUSETTS.

Hon. COMMISSIONER OF PATENTS,
*Washington, D. C.*

SIR: In response to Examiner's action of July 28, 1944, please amend the above entitled application as follows:

Cancel claims 16 and 17

Please add the following claims:

20. A polyalkylene polyamine having each of the two hydrogen atoms on each terminal nitrogen atom replaced by a $-CH_2COOX$ radical, and each of the single hydrogen atoms on each nitrogen atom which is intermediate two alkylene groups replaced by a $-CH_2COOX$ radical, X being selected from the group consisting of hydrogen and alkali metals.

20. A polyalkylene polyamine having each of the two hydrogen atoms on each terminal nitrogen atom replaced by a $-CH_2COONa$ radical, and each of the single hydrogen atoms on each nitrogen atom which is intermediate two alkylene groups replaced by a $-CH_2COONa$ radical.

21. A polyalkylene polyamine having each of the two hydrogen atoms on each terminal nitrogen atom replaced by a $-CH_2COOH$ radical, and each of the single hydrogen atoms on each nitrogen atom which is intermediate two alkylene groups replaced by a $-CH_2COOH$ radical—

## Remarks

Claims 1–7, 10 and 11 are rejected on Polstorff in view of either the French patent 730,760 or the German patent 673,590. Polstorff in view of Dutch patent 46,892 or British patent 490,384 were also cited as cause for rejecting the above claims.

Claim 1 of applicant recites a process wherein the following steps must be followed:

(1) Reacting an *aliphatic amine* with an *alkali metal cyanide* and a *formaldehyde-yielding substance*.

(2) under such alkaline conditions that substantially no hydrolysis of the *cyanide* takes place.

(3) free cyanide present at any time is limited to ¼ of the total amount necessary to react.

(4) free formaldehyde-yielding substance is kept below free cyanide present until the reaction is substantially completed.

(5) removal of ammonia as soon as formed.

It is noted that none of the references alone or in combination show such conditions or intimate that such conditions are either necessary or desirable.

Polstorff does not react the same mixture since no amine is present. Polstorff teaches that formaldehyde will react with potassium cyanide either in dilute or concentrated solutions, and either in the cold or at elevated temperatures.

Polstorff first reacts KCN and $CH_2O$. Applicant takes steps to *prevent* this reaction taking place. Applicant carried out Polstorff's method and found that his reaction is so violent as to be impractical and hard to control—in fact a dangerous explosion occurred when trying it. This statement is in conformity with Polstorff's observations as recorded in the first sentence of the reference.

A further disadvantage of Polstorff's reaction is the production of dark colored substances unless very dilute solutions are used. This makes for varying, though always poor, yields.

By varying the conditions of Polstorff various products are formed. *None* of the five conditions of the first claim as enumerated above are complied with in the Polstorff reference. Polstorff indicates that in very dilute solutions the nitrile formed by the reaction of KCN on $CH_2O$ does not hydrolyze rapidly. Applicant's claim refers to the alkalinity necessary to prevent the hydrolysis of the alkali metal salts used; the hydrolysis of these compounds is too well known and the pH at which they must be maintained is also too well known to require discussion. The fact is in applicant's process if any nitrile is *formed* it must hydrolyze in order to obtain his final product. The hydrolysis of the alkali metal cyanide is prevented, to reduce unwanted side reactions—a necessary step not discovered by Polstorff—or any of the other references.

On the subject of hydrolysis the following may be said. At least two types of hydrolysis may occur. Applicant refers to one; examiner to another. Applicant refers to the hydrolysis of an alkali metal cyanide whose hydrolysis may be written as a mass action equation and is reversible as follows:

$$\frac{[HCN][OH^-]}{[CN^-][H_2O]} = K$$

Examiner refers to another type of hydrolysis which is catalyzed by either $H^+$ or $OH^-$ ions and is substantially irreversible. It is in effect a chemical reaction whose rate of reaction depends upon the

concentration of $H^+$ or $OH^-$ ions. It may be written as follows:

$$RCN + H_2O + MOH \rightarrow RCOOM + NH_3$$
(alkaline solution)

or $$RCN + 2H_2O + HX \rightarrow RCOOH + NH_4X$$
(acid solution)

It so happens that high alkalinity discourages hydrolysis of the alkali metal cyanide and encourages hydrolysis in organic nitriles. This is precisely what applicant desires, i. e. repression of the hydrolysis of the metal cyanide but immediate hydrolysis of any nitrile formed. Acid conditions would, of course, promote hydrolysis of both the metal cyanide and the organic nitrile.

The French reference does not comply with any of the five steps set forth in Claim 1.

The French reference shows at least a two-step reaction under acid conditions with no restrictions on the rate of addition of the materials and no provision for the removal of ammonia as the reaction proceeds.

At best the French reference teaches that an amine and formaldehyde will react and the reaction product will react with HCN. There is no teaching of applicant's process involving entirely different conditions.

The German reference fulfills only one of the five conditions laid down by Claim 1. In the first place different starting materials are used, alkaline conditions are not observed, the rate of cyanide addition is not controlled and ammonia is not removed as formed. The formaldehyde is, however, "added gradually." Certainly it cannot be said that the German reference either in conjunction with Polstorff or alone teaches the process of applicant.

The reason for adding the materials in the particular way set forth by Claim 1 is that a smooth reaction

occurs giving yields greater than 80% based upon the amine used. The process is now operating on a commercial scale to produce the product at the rate of several tons a week—an accomplishment which could not be duplicated from the references cited.

The Dutch patent employs chloracetic acid as does British patent 490,384. Applicant fails to see why this entirely different method of making certain amino polycarboxylic acid compounds should be cited.

The above discussion is with reference to Claim 1, but it applies also to the succeeding process claims inasmuch as they contain the same limitations and others as well. Thus, Claim 2 recites specific conditions involving a vacuum—not shown by the references. Claim 3 employs an excess of formaldehyde added at the end of the reaction—a step not shown by the references. Claims 4, 5 and 6 are limited as to particular compounds or groups of compounds. Claim 7 contains a more precise statement of conditions.

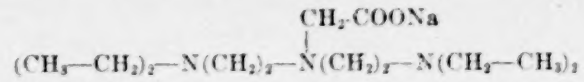
It is submitted that the references cited neither alone nor in any conceivable combination teach the method of applicant as disclosed in Claims 1 to 7 inclusive, 10 and 11.

Claims 14 to 17 inclusive are rejected as unpatentable over the Dutch, British or Fick et al. German patents.

The reaction between diethylene triamine and chloracetic acid is indicated in the Dutch and British patents cited. It is not apparent to applicant that diethylenetriamine reacts with chloracetic acid to give a penta-alkali-metal salt (Claim 14) or the acid (Claim 15) or having all its original amino and imino hydrogens replaced as the salt or acid (Claims 20, 21, formerly Claims 16, 17 respectively). Ex-

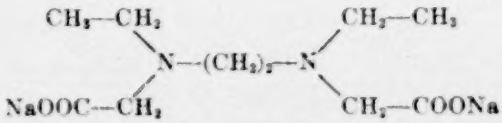
aminer states unequivocally that such a reaction would produce applicant's products. This is contrary to applicant's experimental work which indicates that the imino hydrogens are replaced only with difficulty, if at all, by the chloracetic acid method (see p. 9, line 7 of the specification). Inasmuch as nothing appears in applicant's translation of the Dutch patent to show the reaction of the imino hydrogen, applicant would like to know the Examiner's basis for his statement.

The German patent describes the formation of



in Example 3.

Example 1 shows:



Claim 14 is to a penta-alkali-metal salt. No such compound appears in the reference; neither does any pentaacetic acid as claimed in Claim 15. Claims 16 and 17 which the Examiner considered to be too broad are now rewritten as Claims 20 and 21 to cover the condition that no amino or imino hydrogens are substituted by alkyl groups.

These two new claims are intended to cover the same ground which applicant intended to cover in Claims 16 and 17, but as reworded they are believed to be clear of ambiguity and not too broad. New Claim 19 is generic to 20 and 21 and uses similar language.

Claim 1 is generic to all claims in the case; claims 20 and 21 (replacing 16 and 17) being subgeneric and claims 14 and 15 being species under 20 and 21 respectively. New claim 19 is generic to 20 and 21 and

hence to all the product claims. Non-elected species are retained pending final decision on the allowability of the generic claims.

In view of the foregoing remarks, and the amendments presented herewith, it is believed that this application is in condition for allowance. Reconsideration and allowance are accordingly respectfully solicited.

Respectfully,

FREDERICK C. BERSWORTH,
By FRANK N. HOUGHTON,
*His Agent.*

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Richmond 20, Virginia

Div. 6, Room 272C — Paper No. 7

Address only: The Commissioner of Patents, and not any official by name.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the examiner in charge of this application. Conway P. Coe, Commissioner of Patents.

Applicant: Frederick C. Bersworth. Ser. No.: 491669. Filed: June 21, 1943. For: Aliphatic Polycarboxylic Amino Acids and Process of Making Them.

FRANK N. HOUGHTON,
*35 East Street, Elmwood, Mass.*

Responsive to amendment filed Jan. 3, 1945.

Claims 14, 15, 19, 20, and 21 are again rejected as being unpatentable over the Dutch, British or Fick et al. patents of record. Example 5 of the Fick et al. patent notes that up to five acid residues can be

attached to diethylenetriamine. The patent discloses by example the same materials as applicant has illustrated by example and notes the possible extension to five acid residues. Applicant's disclosure is no more complete, since it discloses the elected subject matter in the same manner. The reference is therefore considered to teach the compounds as well as applicant's descriptions. Applicant has presented no examples teaching the elected compounds and characteristics.

Claims 1 to 7 inclusive, 10 and 11 are again rejected for reasons of record. The claims fail to point out the full method described in the specifications, and fails to state clearly a complete process. The claims, for example, call for adding only one quarter of the cyanide, and less formaldehyde, necessary to complete the reaction. This would only prepare the mono acetic acid or partially convert the raw materials. Apparently the subsequent incremental additions are omitted. The claims must state clearly and concisely a full process. In the absence of pointing out a process which would achieve better results than the references, they are considered to anticipate the claims.

Claims 11, 15, and 21 are rejected as failing to read on the elected species.

O. F. STEARNS,
*Acting Examiner.*

In re Application of Frederick C. Bersworth

Division 6, Room 272–C, Elmwood, Massachusetts

Serial Number 491,669. Filed June 21, 1943. Aliphatic Polycarboxylic Amino Acids and Process of Making Them.

Hon. Commissioner of Patents,
*Washington, D. C.*

Sir: The Examiner in his Action of June 8 1945 has rejected product claims 14, 15, 19, 20 and 21 on the basis of Fick et al, German patent 673,590, with the statement that "Example 5 of the Fick et al patent notes that up to five acid residues can be attached to diethylenetriamine. The patent discloses by example the same materials as applicant has illustrated by example and notes the possible extension to five acid residues." Inasmuch as the said Fick et al patent does not contain such an example or statement, applicant requests that the record be clarified in this regard so that he can make a suitable response to the said Office Action.

Respectfully,

By Frank N. Houghton,
*Agent.*

Department of Commerce
United States Patent Office
Richmond 20, Virginia

Div. 6, Room 272C Paper No. 9

Address only: The Commissioner of Patents, and not any official by name.

All communications respecting this application should give the serial number, date of filing, and name of the applicant.

Please find below a communication from the EXAMINER in charge of this application. Leslie Frazer, Commissioner of Patents.

Applicant: Frederick C. Bersworth. Ser. No. 491669. Filed June 21, 1943. For Aliphatic Polycarboxylic Amino Acids and Process of Making Them.

FRANK N. HOUGHTON
*35 East Street, Elmwood, Mass.*

SUPPLEMENTAL ACTION

Responsive to communication filed June 20, 1945, the following additional references are made of record.

Fick et al (Ger.)—638071, Nov. 9, 1936, 260–464.
Ulrich et al.—2205995, June 25, 1940, 260–464.

The rejection of the first paragraph of the last Office Action (June 8, 1945) is withdrawn in view of applicant's letter of June 20, 1945. The reference was to another Fick et al patent, cited above, which became confused with that of record. It is regretted this error occurred.

Claims 14, 15, 19, 20 and 21 are rejected as being unpatentable over the Dutch, British or Fick et al patents of record or cited above, especially in view of the Ulrich et al patent cited above. Example 5 of the Fick et al patent (#638071) notes that up to five acid residues can be attached to diethylenetriamine. The patent also discloses the acids, salts and corresponding nitriles as a general relationship, and discloses by example the same materials as applicant has illustrated by example noting the possible extension to five acid residues. The Ulrich et al patent includes the formula of the corresponding nitriles. Applicant's disclosure is no more complete, since it

discloses the elected subject matter in the same manner. The reference is therefore considered to teach the compounds as well as applicant's description. Applicant has presented no examples teaching the elected compounds and characteristics.

The last paragraph of the last Office Action is repeated. The rejections are emphasized by the above cited patents.

The six months period for response runs from the date of this letter.

E. W. GENIESSE,
*Examiner.*

*In The United States Patent Office*

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Division 6, Room 272-C
Elmwood, Massachusetts

Serial No. 491,669. Filed June 21, 1943. Aliphatic Carboxylic Amino Acids and Process of Making Them.

HON. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: In response to Examiner's Actions of June 8 and 25, 1945, please amend the above-entitled application as follows:

Claims 1 and 3, line 6 of each, after "cyanide" and before the comma insert —and in total amounts such that for each such replaceable hydrogen atom one molecule of said cyanide and one molecule of said formaldehyde-yielding substance are added during the reaction— line 8 of each, after "mixture" insert —at any one time— line 10 of each, after "present" insert —at any one time—

Claim 1, line 14 cancel "." and add —at any time during the reaction, and finally, after all the cyanide

has been added, adding at least sufficient formaldehyde-yielding substance to react with any unreacted cyanide.—

Claim 3, line 14, after "therein" and before the comma insert —at any time during the reaction—.

Claim 7, line 3, cancel "with" (second occurrence) and insert —in the presence of —. line 14, after "reacted," insert, —said sodium cyanide being added until four mols thereof have been added for each mol of ethylene diamine and the formaldehyde being added— lines 15 and 16, cancel "amount of formaldehyde has" and insert, —four mols have—

Please add the following claim:

10. The process of preparing aliphatic polycarboxylic amino acids which comprises reacting an aliphatic amine having at least one replaceable hydrogen atom attached directly to an amino nitrogen atom, with an alkali metal cyanide and a formaldehyde-yielding substance under conditions sufficiently alkaline that there is substantially no hydrolysis of the said cyanide, while maintaining the amount of said free cyanide present in the reaction mixture at not more than about one quarter of the amount necessary to react completely with the original amine, and maintaining the amount of free formaldehyde-yielding substance present at less than one mol thereof per mol of said free cyanide present and continuing to add said cyanide and said formaldehyde-yielding substance under said conditions until each said replaceable hydrogen atom has been replaced by the radical $-CH_2COOM$ wherein M is an alkali metal, and removing ammonia from the reaction mixture so that substantially no free ammonia is present therein at any time during the reaction, and finally, after all the cyanide has been added, adding a slight excess of for-

maldehyde-yielding substance over that necessary to react with any unreacted cyanide.

Please cancel claims 14, 15, 19, 20 and 21.

## Remarks

Claims 14, 15, 19, 20 and 21 which were rejected have been cancelled.

New claim 22 has been added which defines the metal salt formed by the process in a different but precise manner.

Claims 1 to 7, 10 and 11 were rejected as failing to state a complete process. In view of the apparent ambiguity in this regard, these claims have now been amended to state more clearly applicant's process. The claims are now drawn to point out that all the replaceable hydrogen atoms of the amine are replaced, and that all the cyanide and all the formaldehyde or formaldehyde-yielding substance necessary to do so are added. In view of the amendments submitted herewith, the claims are believed to be allowable.

The references Fick et al., Ulrich, et al., Polstorff et al., the Dutch patent, the British patent, and the French patent are not believed to anticipate the claims as now amended.

Reconsideration and allowance are accordingly solicited.

Respectfully,

Frederick C. Bersworth,
By Frank N. Houghton,
*His Agent.*

## Title Report

No. 491669 C. P.

Name: Frederick C. Bersworth.

The title appears from the assignment records to be vested in: The Martin Dennis Company, Newark,

N. J., a corp. of N. J. Examined up to and including Nov. 2, 1945. This certificate dated Dec. 18, 1945.

H. C. SPIELMAN,
*Chief of Assignment Division.*

No further assignments appear to have been received for record including 43.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Serial No. 491,669

Address only the Commissioner of Patents, Washington, D. C.

DECEMBER 1945.

FREDERICK C. BERSWORTH,
*Assor. Richmond, Va.*

Your application for a patent for an improvement in Aliphatic Polycarboxylic Amino Acids and Process of Making Them filed June 21, 1943 has been examined and allowed with 10 claims.

The final fee, thirty dollars ($30), with one dollar ($1) additional for each claim allowed in excess of 20 (no less sum can be accepted), must be paid not later than 6 months from the date of this present notice of allowance. If the final fee be not paid within that period, the patent will be withheld; *see Revised Statutes, Section 4885 as Amended by Act of Congress Approved August 9, 1939.*

The office delivers patents upon the day of their date, on which date their term begins to run. The preparation of the patent for final signing and sealing will require about 4 weeks, and such work will not be begun until after payment of the necessary final fee.

When the final fee is paid, there should also be sent, distinctly and plainly written, the name of the in-

ventor, title of the invention, and serial number as above given, date of allowance (which is the date of this circular), date of filing, and, if assigned, the names of the assignees.

If it is desired to have the patent issue to an assignee or assignees, an assignment containing a request to that effect, together with the FEE for recording the same, must be filed in this office on or before the date of payment of the final fee.

The final fee will NOT be received from other than the applicant, his assignee or attorney, or a party in interest as shown by the records of the Patent Office.

Respectfully,

CASPER W. OOMS,
*Commissioner of Patents.*

FRANK N. HOUGHTON,
*35 East St., Elmwood, Mass.*

Uncertified checks will not be accepted.

In the United States Patent Office
Division 6—Room 272

IN RE APPLICATION OF FREDERICK C. BERSWORTH

Serial No. 491,669. Filed June 21, 1943. Aliphatic Carboxylic Amino Acids and Process of Making Them.

SUPPLEMENTAL OATH

COMMONWEALTH OF MASSACHUSETTS,
*County of Middlesex, ss:*

Frederick C. Bersworth, the applicant named in the above-entitled application, being duly sworn, deposes and says:

That he is informed and believes that claims of invention have been allowed in the above-entitled application as follows:

1. (1) The process of preparing aliphatic polycarboxylic amino acids which comprises reacting an aliphatic amine having at least one replaceable hydrogen atom attached directly to an amino nitrogen atom, with an alkali metal cyanide and a formaldehyde-yielding substance under conditions sufficiently alkaline that there is substantially no hydrolysis of the said cyanide and in total amounts such that for each such replaceable hydrogen atom one molecule of said cyanide and one molecule of said formaldehyde are added during the reaction, while maintaining the amount of said free cyanide present in the reaction mixture at any one time at not more than about one quarter of the amount necessary to react completely with the original amine, and maintaining the amount of free formaldehyde-yielding substance present at any one time at less than one mol thereof per mol of said free cyanide present until substantial

completion of the reaction of said amine has been effected, and removing ammonia from the reaction mixtures so that substantially no free ammonia is present therein at any time during the reaction, and finally, after all the cyanide has been added, adding at least sufficient formaldehyde-yielding substance to react with any unreacted cyanide.

2. (2) The process according to Claim 1 wherein the reaction is carried out at a temperature of between about 60° C. and about 80° C. and at a pressure such that the reaction mixture is substantially at the boiling point at said temperature, until the major part of the reaction has been completed, and the temperature and pressure are then gradually raised while maintaining substantially boiling conditions, until completion of the reaction.

3. (3) The process of preparing aliphatic polycarboxylic amino acids which comprises reacting an aliphatic amine having at least one replaceable hydrogen atom attached directly to an amino nitrogen atom, with an alkali metal cyanide and a formaldehyde-yielding substance under conditions sufficiently alkaline that there is substantially no hydrolysis of the said cyanide and in total amounts such that for each such replaceable hydrogen atom one molecule of said cyanide and one molecule of said formaldehyde are added during the reaction, while maintaining the amount of said free cyanide present in the reaction mixture at any one time at not more than about one quarter of the amount necessary to react completely with the original amine, and maintaining the amount of free formaldehyde-yielding substance present at any one time at less than one mol thereof per mol of said free cyanide present until substantial completion of the reaction of said amine has been effected, and removing ammonia from the reaction mixture so that

substantially no free ammonia is present therein at any time during the reaction and finally, after all of the cyanide has been added, adding a slight excess of formaldehyde-yielding substance over that necessary to react with any unreacted cyanide.

4. (4) The process according to Claim 1 wherein the formaldehyde-yielding substance is formaldehyde.

5. (5) The process according to Claim 1 wherein the amine is a primary aliphatic amine.

6. (6) The process according to Claim 1 wherein the amine is a primary aliphatic diamine.

7. (7) The process of preparing the sodium salts of ethylene diamine tetraacetic acid which comprises reacting ethylene diamine with sodium cyanide and formaldehyde in the presence of sufficient sodium hydroxide to bring the pH to at least about 9 while maintaining a temperature of between about 60° C. and about 80° C. and a pressure such that the reaction mixture is substantially at the boiling point at said temperature, and maintaining the amount of free cyanide present in the reaction mixture at any one time at not more than one quarter of the amount necessary to react completely with the ethylene diamine, and maintaining the amount of free formaldehyde present in the reaction mixture at any one time at about five per cent less than the amount of sodium cyanide present, which five per cent is based on the total quantity of sodium cyanide to be reacted, said sodium cyanide being added until four mols thereof have been added for each mol of ethylene diamine and the formaldehyde being added until at least about 95 per cent of the theoretically required four mols have been added, adding sufficient additional formaldehyde substantially completely to react with the unreacted sodium cyanide, increasing the pressure and heating the solution to boiling at a

temperature at least about 110–120° C., and removing ammonia from the reaction mixture so that substantially no free ammonia is present therein at any time.

10. (8) The process according to Claim 1 wherein the amine is diethylene triamine.

11. (9) The process according to Claim 1 wherein the amine is n-butylamine.

22. (10) The process of preparing aliphatic polycarboxylic amino acids which comprises reacting an aliphatic amine having at least one replaceable hydrogen atom attached directly to an amino nitrogen atom, with an alkali metal cyanide and a formaldehyde-yielding substance under conditions sufficiently alkaline that there is substantially no hydrolysis of the said cyanide, while maintaining the amount of said free cyanide present in the reaction mixture at not more than about one quarter of the amount necessary to react completely with the original amine, and maintaining the amount of free formaldehyde-yielding substance present at less than one mol thereof per mol of said free cyanide present and continuing to add said cyanide and said formaldehyde-yielding substance under said conditions until each said replaceable hydrogen atom has been replaced by the radical $-CH_2COOM$ wherein M is an alkali metal, and removing ammonia from the reaction mixture so that substantially no free ammonia is present therein at any time during the reaction, and finally, after all the cyanide has been added, adding a slight excess of formaldehyde-yielding substance over that necessary to react with any unreacted cyanide.

That he verily believes himself to be and to have been, prior to the filing of the said application, the original, first and sole inventor of the inventions claimed in and by the aforesaid claims; that he does

not know and does not believe that the same was ever known or used before his invention or discovery thereof, or patented or described in any printed publication in any country before his invention or discovery thereof, or more than one year prior to the aforesaid application, or in public use or on sale in the United States for more than one year prior to the said application and that the same has not been abandoned; that said inventions have not been patented in any country foreign to the United States on an application filed by him or his legal representatives or assigns more than twelve months prior to the said application, and that no application for patent on said inventions has been filed by him or his representatives or assigns in any country foreign to the United States prior to the said application.

FREDERICK C. BERSWORTH.

Sworn to and subscribed before me this 14th day of January 1946.

[SEAL] ALLAN L. SPURR.

My Commission expires Nov. 14, 1952.

FRANK N. HOUGHTON
PATENT AGENT
35 EAST STREET, ELMWOOD, MASSACHUSETTS.

*June 18, 1946.*

Hon. COMMISSIONER OF PATENTS,
*Washington 25, D. C.*

SIR: I am enclosing check for thirty dollars ($30.00) to cover the final fee in the following application:

Applicant: Frederick C. Bersworth. Serial No.: 491,669. Filed: June 21, 1943. For: Aliphatic Polycarboxylic Amino Acids and Process of Making them. Allowed: December 26, 1945. Assignee: This was originally assigned to The Martin-Dennis Company,

Newark, N. J., but I understand has recently been reassigned by them back to the applicant.

I also request that you delay three months beyond the date of receipt hereof, as permitted, in issuing the patent on this application.

Yours very truly,

FRANK N. HOUGHTON,
*Agent for Applicant.*

Enclosure—check.

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE

RICHMOND, VIRGINIA, *June 24, 1946.*

Applicant: Bersworth, F. C. Serial No.: 491669. Filed: June 21, 1943. For: Aliphatic polycaroxylic amino acids and process of making them. Final Fee: June 19, 1946. Issue Date: Sept. 17, 1946.

FRANK N. HOUGHTON,
*35 East St., Elmwood, Mass.*

SIR: Your letter requesting deferment under Section 4885 R. S. has been received

Respectfully,

J. A. Brearley,
J. A. BREARLEY,
*Chief Clerk.*

bg.

NUMBER (Series of 1929)

491669

PATENT No. 2407645

1943

DATED SEP 17 1946

DIV. 6

(EX'R'S BOOK) 89-89-47

Name FREDERICK C. BERSWORTH,

Assor. to The Marten Dennis Company, of Newark, N. J., a Corp. of New Jersey

of VERONA

State of NEW JERSEY

Invention ALIPHATIC POLYCARBOXYLIC AMINO ACIDS AND PROCESS OF MAKING THEM

ORIGINAL

APPLICATION FILED COMPLETE JUNE 21, 1943

PARTS OF APPLICATION FILED

Petition, Specification, Oath, First Fee $30 — JUNE 21, 1943

Examined and passed for Issue DEC 26 1945, 19 — Ex. Div. 6

Notice of Allowance DEC 26 1945, 19

Final Fee $30 June 19, 46, 19

AGENT FRANK N. HOUGHTON 35 EAST ST ELMWOOD MASS.

Associate Attorney

No. of Claims Allowed 10 Print Claim 1 In O. G. Class 260-534

Title as allowed Aliphatic Polycarboxylic Amino Acids and Process of Making Them

Division of App., No. , 19 , filed

6/21/43 mk

PAPERS RE
JUN 28 1943
DIVISION 6

260-534

# 1943

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